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ESSENTIAL OILS
&
AROMATIC CHEMICALS

A SYMPOSIUM HELD AT DEHRA DUN
OCTOBER 6-9, 1955
UNDER JOINT AUSPICES OF
CSIR AND THE FOREST RESEARCH INSTITUTE
AND COLLEGES

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PREFACE

The avowed object of the various Research Committees functioning under the Council of Scientific & Industrial Research (CSIR), besides sanctioning grants-in-aid for research—scientific and technical—bearing on the subjects with which they are connected, is to place before the public a graphic account of the results of their work. Holding of symposia, seminars and conferences is a popular form of publicizing the aims, activities and achievements of the Research Committees. The need for holding a symposium to focus attention on the work of the Essential Oils Research Committee since its inception 15 years ago, was long felt. With the approval of the Governing Body of the CSIR, a symposium on *Research & Development of Essential Oils & Aromatic Chemicals* was held at the Forest Research Institute, Dehra Dun, during October 6-9, 1955 under the joint auspices of the Essential Oils Research Committee and the Forest Research Institute & Colleges.

The fact that the symposium was attended by about 100 delegates from all over India, representing various Departments of the Government of India, State Governments, research laboratories, private firms and persons employed in the culture and processing of aromatic raw materials and manufacture of finished products, such as soaps, perfumes, cosmetic and toilet articles, bears ample testimony to the utility of such organized effort to give an overall picture of the progress of work in the field of essential oils and aromatic chemicals and the necessity for taking active steps for their further development.

A number of original papers dealing with indigenous essential oils, their composition, uses and commercial possibilities were contributed by the delegates and elicited useful comments and helpful criticism.

Grateful thanks are due to Dr. Sadgopal for taking the trouble of collecting and compiling the papers contributed for inclusion in the symposium, and to the CSIR for undertaking the publication.

A. K. Menon
Chairman
Essential Oils Research Committee

Kozhikode
Oct. 15, 1955

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Welcome Address

SHRI K. L. AGGARWAL
President, Forest Research Institute & Colleges, Dehra Dun

I deem it a privilege to extend to you all a hearty welcome to the Forest Research Institute on the occasion of the first Symposium on Essential Oils and Aromatic Chemicals in India. I am particularly grateful to Dr. Deshmukh for finding time, in the midst of his pressing engagements, to inaugurate this symposium and to Prof. Thacker who braved all obstacles on the road and squeezed in time to come to the Forest Research Institute for this function. This symposium, as you are all aware, was scheduled to be held during the last year but had to be postponed due to unavoidable reasons. I am glad that after all it has been possible to meet the wishes of the Essential Oils Research Committee of the Council of Scientific & Industrial Research for holding the symposium at this time.

I have no doubt that the reason why the Essential Oils Research Committee was anxious to give to this Institute the honour of organizing this first symposium is the pioneer role played by the Forest Research Institute, since its inception in 1906, towards the intensive development of natural essential oils in India. It is needless to point out that almost all the important essential oils of India, with the exception of a few like rose *attars*, are the products obtained from Indian forests. As such, this Institute was bound to play a vital part in their development. My predecessor, Shri C. R. Ranganathan, now the Inspector-General of Forests to the Government of India, readily agreed to organize the symposium at this Institute ; but on account of certain difficulties the work could not be taken up in earnest until some 4 months ago which made it an uphill task as the preparations for the symposium had to be started almost from scratch. It was mainly because of the unfailing confidence reposed in this Institute by the Essential Oils Research Committee headed by its seasoned energetic Chairman, Shri A. K. Menon, and also the continuous hard work put in by its Secretary, that this work could be planned and executed in a systematic manner within the limited time at our disposal.

Fortunately, most of the equipment for processing the plant materials was already available in the Chemistry of Forest Products Branch of this Institute though it had been lying unused for some years. We had also received under the Technical Co-operation Administration solvent extraction and fractional distillation plants which would make the equipment up-to-date. The symposium enthused us to go ahead with the erection work, and plans for putting up an assembly in keeping with the status of this Institute were worked out. Even then it would have been impossible to complete this task in such a short time but for the willing and active collaboration of the Officer-in-charge of the Service Branch and the members of the staff deputed to do this job in the various sections, all of whom worked wholeheartedly for remodelling and re-erecting the entire chemical equipment. The Central Public Works Department did their part in re-laying the floors and putting up electric and other fittings. This equipment, worth more than a lakh of rupees, has been fully renovated and erected in our own workshops. With further improvements, which we are hoping to effect, I am sure this laboratory will become one of the best equipped laboratories in the East.

A visit to this laboratory will give you a glimpse of our future plans of work under the second Five-Year Plan. I can assure everyone interested in this field of work that the Forest Research Institute is determined to play its fullest part.

When material regarding the contributions made in this field was collected, we found with some surprise, not unmixed with pride, that the contributions by the scientific workers of this Institute were not only the largest in the field but also very important. The monumental work of Puran Singh, Simonsen, Gopala Rau, Krishna and others accomplished in the chemical laboratories of this Institute has made its name world-famous. It is no wonder that the wealth of scientific literature accumulated on the vast aromatic resources of India in this Institute for 50 years has always been looked upon as the treasure house of scientific information on essential oils. It was in 1950 that under the reorganization scheme of the Institute, a start could again be made for strengthening the research on and the development of natural essential oils in the Chemistry of Forest Products Branch. It naturally took some time to come to grips with the various problems by collecting once more the broken threads of past work and achievements in a systematic manner and bringing them to a point wherefrom further work could be planned with confidence. As a result of this, within the short period of the last five years, the Chemistry of Forest Products Branch has made some very useful and commercially important advances in the field of essential oils. To mention a few cases, I would like to draw your attention to the development of economic methods for the extraction of cent per cent pure essential oils and *attars* of agar-wood, costus roots, homalomena roots yielding an oil with about 40-45 per cent of linalool and Himalayan cedar-wood oil from deodar wood waste. The introduction of rotational system of extraction of vetiver roots (available in Bharatpur forest areas) and utilization of the roots for the production of better quality and better yield of vetiver oil is already reported to have raised the price of *khus* in Bharatpur by about 50 per cent this year. The development of a cheap mobile field unit giving very economic yields of essential oils with the aid of the Forest Research Institute condenser, development of an essential oil trap and evolution of an economic process for the sublimation of crude camphor into chemically pure camphor are the other important contributions from this Branch. The utilization of decamphorized ocimum oil as an effective solvent for gold lustering of glass and ceramicware and standardization of a process for distilling a high grade pharmaceutical oil of chenopodium have also been perfected. Work is also in progress on the equally important cultural aspects of essential oil bearing plants in the Minor Forest Products Branch of this Institute.

The equipment that has been installed will now enable us to push forward research and development in the field of natural essential oils at this Institute. Apart from these direct steps, I am happy to note that by rendering scientific aid and service to workers and institutes in different parts of India, this Institute has been able to create in various centres a spirit of mutual co-operation and fellowship. It is highly satisfactory to find here today the top ranking workers from every corner of the country representing not only the progressive elements of commerce and industry but also the Universities of Lucknow, Bombay and Aligarh and leading laboratories like the Indian Institute of Science, National Chemical Laboratory of India, Harcourt Butler Technological Institute, Drug Research Laboratory, Central Food Technological Research Institute, National Botanic Gardens and many others. By looking at this representative gathering I feel confident that this long neglected but important dollar-earning industry of essential oils is destined to play an increasingly important role in developing India's natural resources. From this Institute I assure you all the fullest co-operation and scientific assistance.

The Essential Oils Research Committee and workers from all over India expressed a keen desire that along with this symposium, an exhibition of essential oils and related products and the industries served thereby should also be organized. You will have an opportunity to witness this afternoon the result of a month's labour in this direction. I hope you will find here a representative and varied collection of exhibits which will form a potential nucleus of a permanent museum of the industry that we are planning to set up in this Institute in the near future.

It is really a matter of pride for me to welcome to this Institute the delegates numbering over 80, representing all those interests which are concerned with research and development in the field of essential oils. The spontaneity with which progressive industrial houses have responded to the request of the Secretary for social receptions to the delegates attending the symposium is very heartening indeed. What is most striking is the fact that about 70 original scientific and technical papers have been received at such a short notice. I am further given to understand that a good many papers are of an outstanding nature.

Another important feature of this symposium is that three leading organizations interested in the development of the essential oils industry are going to meet here simultaneously during this week. I am referring to the Essential Oils Research Committee of the Council of Scientific & Industrial Research, the Essential Oils Sectional Committee of the Indian Standards Institution and the Scientific Research Committee of the U.P. Government. The work of the Essential Oils Research Committee needs no introduction and I will not, therefore, refer further to its creditable achievements. The Scientific Research Committee of U.P. has taken commendable initiative in financing research schemes on roses, vetiver and valerian oils. The fact that this Committee is meeting here shows its great interest in the development of the essential oils industry. The work and achievements of the Essential Oils Sectional Committee of the Indian Standards Institution are also very well known. I am happy to find that the contributions made by the Forest Research Institute during the last five years for the development of Indian Standard Specifications and Methods of Tests have been very well received and are influencing the course of developments at the level of the International Standards Organization of which India is a member-country. It is no mean achievement of the Indian Standards Institution that about one dozen Indian standards have been released and put in operation since 1950. The Forest Research Institute has been requested by the Indian Standards Institution to act as the National Referee Testing Laboratory by maintaining the permanent standards for gum rosin and issuing the secondary standards for general use and reference. I need not repeat the assurance extended to the Indian Standards Institution by my distinguished predecessor, Shri C. R. Ranganathan, that the Forest Research Institute shall always keep aloft the flag of Indian standards.

To the members of all the three committees, I extend a hearty welcome and wish them all success in their important deliberations.

I may now express my deep appreciation at the large number of highly encouraging messages of goodwill received by us for the success of this symposium including those from Dr. S. Radhakrishnan, Vice-President of the Indian Union, Maulana Abul Kalam Azad, Union Minister for Natural Resources & Scientific Research and Education, Shri K. M. Munshi, Rajyapal of U.P. and various distinguished scientists from India and abroad. This is a very good augury for the success of the symposium.

Before I conclude I wish to add that the Forest Research Institute is situated in a world of its own known as the 'New Forest'. We have the advantage of having been placed in beautiful surroundings at the feet of the Himalayas. This, however, deprives us of some of the common amenities of life. Consequently, most of you will find your stay perhaps not very comfortable. The accommodation and other services could not be made as satisfactory as we would like to offer but I am confident that you will gladly bear with us for these shortcomings. I also hope you will be able to establish some useful contacts during your stay at this Institute. The various branches, museums, workshops and laboratories of this Institute will be extending to you a hearty welcome. I shall be happy if this union of the active workers in the field of essential oils leads to greater and intensive development of the industry in the near future. In the economy of a country like India, intensive development and utilization of her natural resources is imperative. In this noble effort of yours, the attention not only of India but also of the world is focused on your deliberations. May your co-ordinated efforts lead to greater success and prosperity of this important industry.'

Inaugural Address

DR. P. S. DESHMUKH
Union Minister for Agriculture, New Delhi

I am grateful to you for affording me this opportunity to be in your midst today and am happy to associate myself with this symposium on essential oils. Essential oils present a wide range of variety and properties and are derived from various parts of different plants. They have a very important part to play in many spheres of human activity. One of the best known of the uses of essential oils is, of course, in the manufacture of perfumes and cosmetics. In fact, it has been observed that no page of human history antedates the use of perfumes and cosmetics. The use of perfumes evidently originated with religious observances where fragrant incense was burnt by man from ancient times at the altars of gods. Aromatic substances have also played a prominent part in offerings, for anointing and embalming and for rendering the air in places of worship pleasant-smelling.

There is ample evidence to show that the production and use of perfumes and aromatics had been developed to a considerable extent in our country for a long time. Famous travellers of those days like Fahien have described this country as the land of aromatic flowers, fruits, woods, roots, resins and grasses. Of these perhaps the best known have been sandalwood and its oil, as they have been favourites from times immemorial in the world of perfumes. In fact, there appears to have been a regular barter trade in sandalwood since ancient times when caravans carried the precious wood from India to Egypt, Greece and Rome. India came to be known for her high class perfumes ; and centres like Kanauj, Jaunpur, Ghazipur, Lucknow, Poona and Pandharpur acquired great reputation in the past for their perfumery products.

It appears, however, that in comparatively recent times our country lost this pre-eminent position as the manufacturer of superior perfumes and aromatics, probably because of inefficient organization, malpractices especially by way of adulteration, and failure to advance with the times in our methods of processing. Like many of our old habits our sense of smell has also undergone a change which gradually discarded the *otto-dilbahar* and took to Coty's Chypre. Our manufacturers, however, like the more conservative sections of our society, never imagined that our noses would refuse to smell the strong rose and *khus* and go in for the comparatively soft smelling Parisian perfumes. As a consequence, I understand that we are now exporters of raw materials, namely, essential oils or products yielding essential oils and importers of finished perfumery articles. It should be one of the objectives of this symposium to devise ways and means to help in the restoration of this neglected art to its rightful place in the country and thus save exports of valuable raw materials and even resume exports of finished perfumes, aromatic chemicals and pharmaceuticals based on essential oils.

The use of essential oils, however, is not confined to the preparation of perfumery. Either the essential oils as such or perfumes or other materials processed from them are put to a great variety of uses. Important toilet, medicinal, food and other preparations like soaps, disinfectants insecticides, fungicides, chewing and smoking tobacco, chocolates and toffees and other confectionery, syrups and aerated waters, *agar-battis,* pharmaceutical preparations and even writing pads and invitation cards include the use of suitable essential oils. With such a diversity of uses to which these materials can be put, it is necessary that not only their collection, extraction and processing are placed on an organized footing, but their production also is systematically augmented.

The important essential oils known in the country are oils of sandalwood, lemongrass, palmarosa grass, gingergrass, and vetiver. Essential oils are also derived from various seeds such as ajowan, aniseed, coriander and fennel. Perfumes are also derived from oils from a variety of flowers such as rose, jasmine and various species of citrus.

I am glad to note that observations have been recorded on a fairly extensive scale regarding the resources of the plant kingdom of India in this respect. A number of botanists and pioneers in forestry have drawn attention to our immense potential for aromatic substances. In the field of chemical investigations into these substances also much pioneer work has been done ; it cannot be gainsaid that much remains to be achieved, especially the evolution of modern methods of distillation and further utilization of essential oils. I would suggest that this symposium should indicate lines along which selected plants yielding valuable essential oils could be profitably cultivated in different parts of the country and how the raw material resources of the country could be adequately expanded. At the same time, improved methods of distillation and isolation of essential oils have to be developed. Steps have also to be taken for determining the ways and means for their increased use.

It should be apparent that while many of these products occur in our forests and fields naturally, others require to be cultivated. It is recognized that the incidence of plants yielding essential oils is very scattered in the country and efforts to cultivate them have been made on a very limited scale. There is much scope for development not only by adopting regular methods of conservation and collection, but also by raising plantations or by improving the desired species in the natural crops. Further, it is a recognized fact that till recently foresters concerned themselves mainly with timber production and forest protection and the development of what is popularly referred to as minor forest products has received only secondary attention. In fact there is much scope for gainful development in this field with particular reference to essential oils.

It gives me great pleasure to note that a number of organizations, both governmental and private, are interesting themselves in every field of activity connected with essential oils. The forests of India have been the main source of supply of plant materials yielding essential oils and the forest departments have been concerned with such products as an important item of minor forest produce ; but there is much that could be done by them to intensify and regulate not only the collection of what occurs naturally but improve on it by resorting to systematic planting.

Private efforts to raise plantations of species yielding essential oils, though successful, have been made only on a limited scale up-to-now. I would ask interested individuals attending this symposium to give thought to this question. I have in mind particularly such items as *linaloe* and *patchouli*. The latter has figured prominently in questions in both Houses of Parliament. In the fields of isolation of the essential oils and of their further processing, research centres like the Forest Research Institute and the Indian Institute of Science, Bangalore, have been pioneers and have much good work to their credit. More recently, a number of other organizations, under the able guidance of the Essential Oils Research Committee, Council of Scientific & Industrial Research, (as also other bodies like the Uttar Pradesh Scientific Research Committee) have also begun to take interest in this important work. Quite a few firms have shown commendable enterprise in developing the commercial production and utilization of essential oils, although for one reason or other they have not always been prosperous. It is gratifying to note that some reputed foreign firms established in our country are also evincing active interest in the development of Indian essential oils and their uses.

It is also satisfying to learn that the Indian Standards Institution has taken interest in the formulation of suitable standards for essential oils and that standards have been laid down for sandalwood and other oils. This should ensure that in future this industry will not suffer in the unfortunate manner in which it received a setback due primarily to poor output, adulteration and inefficient methods and processes in the past.

I understand that most of these institutions, bodies and concerns are taking part in this symposium. The symposium should therefore be able to arrive at conclusions which will form the basis for the guidance of all these departments, organizations, firms, institutions and individuals in their future work in the particular fields of activity they may happen to be interested in, so that each may contribute its best towards the overall development of Indian essential oils. I have, however, to draw attention to one particular aspect of such work ; and it is this: While it would be good in the future that a number of institutions and firms should interest themselves in this work, it is necessary that waste by way of duplication of work should be avoided as much as possible. I understand that some steps have recently been taken in this connection at the instance of the Ministry of Natural Resources & Scientific Research. All the same, I would suggest that some form of a permanent co-ordinating organization should be set up to deal with all research and development work in this field in such a way as to foster harmonious development all over the country. I would also draw the attention of the Essential Oils Research Committee, to whose initiative we owe this first symposium, to this point. You will all agree that it is only proper that the Forest Research Institute, Dehra Dun was chosen as the venue for this symposium, as the Institute has been a pioneer in the field of investigation of essential oil bearing plants and of essential oils. In fact, it can be said that the large-scale development of industries manufacturing or utilizing sandalwood, turpentine, eucalyptus and other oils, is primarily due to the early work done in this Institute. Recently, the laboratories dealing with this subject have been remodelled and research facilities improved. I trust you will have an opportunity during the course of your stay here to see these developments for yourselves and to suggest ways and means by which they can be utilized to the maximum advantage.

I am aware that during this symposium you will have ample opportunities to discuss on the basis of the scientific and technical papers to be presented before you, the various subjects of interest in this field and to take stock of the present position in order that you may evolve suitable steps for further development. I wish this symposium and each of you individually all success in your efforts to improve and develop this national resource and the important industries based on it. I hope your visit to the Forest Research Institute on this occasion will prove pleasant and fruitful and that you will return to your own centres of work with happy memories of your stay here.

Address

Shri A. K. MENON

Chairman, Essential Oils Research Committee, CSIR

On behalf of the Essential Oils Research Committee of the Council of Scientific & Industrial Research I offer you all a hearty welcome to this first *Symposium on Research and Development in Indian Essential Oils and Aromatic Chemicals*. To you, Dr. P. S. Deshmukh, we are deeply indebted for having so readily consented to inaugurate this symposium and for having taken the trouble to come to Dehra Dun in spite of your numerous and pressing engagements elsewhere and the inclement weather. This is ample proof of the great interest you take in the development of agriculture on which the major part of the population of India is dependent for its livelihood. Aromatic plants which yield essential oils find only a minor place in the agricultural map of India ; but their potentialities are tremendous. It is seen from literature that citrus seeds were taken from India to Italy, and in Sicily, the citrus oil industry has assumed enormous proportions. Similarly vetiver (*khus*) oil which we import from Reunion Islands and other places is extracted from roots of plants, rootstocks of which were originally taken from India. Spices like pepper, ginger, and cardamom which are indigenous to India are now extensively grown in other countries and are competing with our products. It is clear that India has lagged behind. What is needed now to develop the industry connected with the cultivation of essential oil crops, is the application of modern methods, selection of proper seeds, adoption of ideal conditions of cultivation, application of manure and a study of soil structure and climate. We look up to you, sir, for rendering all the necessary help and guidance in agricultural matters pertaining to the cultural side and above all for a proper appraisal of the potential resources of this important agricultural crop.

Professor Thacker, Director, Scientific & Industrial Research, every minute of whose time is packed with important work connected with the administration of the Council of Scientific & Industrial Research, takes a keen interest in research activities and development of our natural resources. We are indeed grateful to you, sir, for your kindly presiding over the opening session and giving us much valuable advice and we expect to receive your continued support and encouragement for developing the essential oils industry. The object of the symposium is to take stock of the achievements that have taken place in the essential oils industry in our country and to focus attention to the need for taking steps towards its further development. The main requirements in India for the rapidly growing field of essential oils are: (*i*) more intensive research—agricultural and chemical ; (*ii*) augmentation of financial aid for such research ; and (*iii*) wider recognition of the advantages of developing the essential oils industry in India.

The Essential Oils Research Committee has played an important role in the attainment of progress in various directions. As a result of the advice and technical help rendered by the Committee, private parties have taken to growing aromatic plants and applying modern methods of extraction of oils and preparing isolates. It is heartening to see such a large number of delegates from all over India, gathered here to participate in the symposium. Quite a number of papers have been received from persons actually engaged in the cultivation of plants, production of oils, and utilization of these oils in various industries. Several research papers of considerable academic and technical value have been contributed by scientific workers in various government institutes and private laboratories and firms. To all those who have contributed these valuable papers we offer our sincere thanks.

Another feature which is going to make the symposium attractive and complete is the exhibition which has been arranged. I am glad to say that there has been full co-operation from the planters, distillers, manufacturers and research laboratories. You would never realize until you see, the vast range of exhibits which have been displayed to evoke your interest and admiration. For the lay visitor, especially the ladies, there are some enchanting essences and scents, toilet soaps, face powders and creams which derive their charm from the subtle essences of flowers and herbs grown in India. For the student of chemistry, research worker, manufacturer and merchant, the exhibition will be an eye-opener to the vast range of aromatic raw materials, distilled essences and extracts, produced in this country and outside. You will be surprised at the variety of uses to which essential oils are put. The lime (*chuna*) you use with your *pan* is scented with rose water, the *pan* or betelnut is agreeably perfumed, while the tobacco you chew is also likewise made aromatic by the help of finely blended perfumes. Foods, beverages and sweets are also made more palatable by the addition of subtle perfumes. Indian *attars* have such a charming perfume that at times some people do away with bathing for days together.

The irresistible attraction of delicately perfumed cosmetic cream or hair oil is well known. Perfumes are also used to mask the smell of inks, polishes, incense, and similar substances. Joss sticks and *agar-battis*, which contain different aromatic ingredients, readily dispel bad odours. In theatres and picture houses, perfumes are sprayed to diffuse a pleasant and refreshing aroma. The real purpose and aim of practical perfumery is to provide means of increasing the enjoyment of life.

To the various exhibitors, I offer our sincere thanks for readily sending their products, notwithstanding the short notice they received.

Besides the exhibits, there are a number of charts which give data of production of essential oil plants grown in India, statistics of exports and other valuable information.

In this connection, I have to make special mention of the splendid work which Dr. Sadgopal, our energetic Organising Secretary, has put in arranging everything in such an efficient way. He and his band of enthusiastic workers have spared no pains and worked day and night. Dr. Sadgopal has several years of intensive research work to his credit in the field of essential oils. He has placed some of his rare collection of literature, specimens of special essential oils, *attars* and such products which have added to the utility of the exhibition.

I should like also to draw your attention to the Essential Oil Processing Laboratory. Here, Dr. Sadgopal has on show an up-to-date distillation plant of considerable capacity side by side with smaller units. This is of great educational value to those contemplating setting up factories for essential oil production. On behalf of all present and on my own behalf, I offer sincere thanks to Dr. Sadgopal and his assistants for all they have done.

Regarding the choice of the Forest Research Institute as the venue for holding the first symposium, I should say that this Institute has the first claim for this signal honour. Work on essential oils formed part of the activities of this Institute from its very beginning about 50 years back. Names of Puran Singh, Simonsen and Krishna stand out as pioneers in this field. I am glad to say that the good work is continued by other workers especially by Dr. Sadgopal. I wish to thank on behalf of the delegates and others Shri Aggarwal, the President of the Institute, for placing the beautiful halls at our disposal and for the excellent facilities provided to the delegates and others.

I will be failing in my duty if I do not refer to the generous gesture of the firms and others in extending hospitality to the delegates. I offer all those concerned in this matter my hearty thanks. Lastly, I have to refer to the distinguished persons who agreed to preside over the symposium sessions. Shri P. A. Narielwala who was primarily responsible for bringing the essential oils industry to the notice of the public and did so much for its growth was to have presided on the second day of the session. He has been obliged to cancel the arrangement owing to unavoidable reasons. I am glad to say that Shri K. L. Aggarwal, President, Forest Research Institute has kindly agreed to fill this void. I thank Dr. Moudgill who readily responded to my request to take the chair on the third day.

Before I conclude I offer my best thanks on behalf of the organizers of the symposium and on my own behalf to fellow delegates and visitors who have come to this distant place at considerable expense and inconvenience.

Chairman's Address

PROF. M. S. THACKER
Director, Scientific & Industrial Research, New Delhi

It gives me great pleasure to be present at such a vast gathering of scientists and industrialists and I consider it a privilege to be the chairman of the symposium.

I am thankful to Hon'ble P. S. Deshmukh personally and on behalf of the Council of Scientific & Industrial Research for his readily agreeing to inaugurate the symposium and for taking the trouble to come over here in spite of his numerous and more pressing engagements elsewhere and the bad weather.

The Council of Scientific & Industrial Research has played a certain part in the organization of this symposium. But more significant than this is the part played by the Forest Research Institute. I am grateful to the President, Forest Research Institute and Colleges, Shri A. K. Menon, Chairman of the Essential Oils Research Committee and Dr. Sadgopal who had taken a lot of trouble in organizing the symposium and I hope that their efforts will be crowned with success. It had been the practice of the Council of Scientific & Industrial Research, ever since its inception, to organize and arrange such seminars and symposia which are not only important from the industrial point of view but also give impetus to further scientific developments.

I do not think it worthwhile to dilate on the scientific work being done in this particular field. The Council of Scientific & Industrial Research have a correct appreciation of the position and had recently convened a conference at Delhi under the auspices of the Essential Oils Research Committee and had taken certain decisions. They had already recommended the establishment of four centres for research on essential oils and aromatic chemicals in the country, to be followed up by more centres if necessary. Shri Roerich has done a lot of work for the development of linaloe plantations in order to increase the output of this important essential oil, so much in demand in India and abroad.

In the programme of work of the Council of Scientific & Industrial Research a large sum of money has been set apart for the improvement of this industry and I have also made a request to the Planning Commission to give money to the tune of one crore of rupees for the development of this industry in the near future.

I am happy to be in this representative gathering of scientists and industrialists from all corners of India and wish all success to the first symposium. I very much appreciate the excellent organization of the Exhibition and also the Essential Oils Processing Laboratory, which, I hope, will be of great interest to all the delegates and visitors.

Perfumes in the National Plan

MADAN LAL JAIN
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The scope of the perfumery industry has been outlined and its present position discussed. Necessity for the systematic cultivation of aromatic plants and standardization of their products has been emphasized. It is suggested that more attention be devoted to research on the utilization of odoriferous essential oils. Foreign co-operation may be helpful in the field of aromatic chemicals, and cultivation and distillation of some particular plant material, but the sphere of synthetic essential oils may be reserved for the indigenous entrepreneurs.

Perfumes constitute an indispensable ingredient of the necessities of daily life. They are essential adjuncts of ice creams, sweets, toffees, drinks, chewing tobaccos, *pan masalas, bidis* and cigarettes, and are added to soaps, hair oils, creams, toilet powders, dentifrices, *agarbattis,* disinfectants, antiseptics and medicines. Indigenous manufacture of perfumes is, therefore, of prime importance.

Perfumes are judicious mixtures of various odoriferous principles blended together for the sake of aesthetic pleasure. The fragrant substances are obtained from flowers, leaves, roots and bark of plants like jasmine, lemongrass, palmarosa, eucalyptus and *khus*, and also from animals such as musk deer and civet cat. In addition to these natural products, the organic chemists have placed at the disposal of the industry an ever expanding array of aromatic chemicals with agreeable odours.

During the last thirty years the Indian perfumery industry in spite of keen competition from well-established foreign interests has steadily developed and acquired a respectable place in the perfumery trade. The attitude of the National Government has been sympathetic towards the industry. But it appears that some modifications in the import policy and tariff have unfortunately failed to afford the intended protection due to some lacunae. Lately the Government raised the duty on perfumery compounds from 31¼ per cent to 66½ per cent *ad valorem,* but exempted the same compounds under the name of synthetic essential oils from higher duty. Aromatic chemicals which form the major constituents for compounding synthetic essential oils, with a few exceptions, are not yet produced in India, and have to be imported. The import duty levied on them is 37⁴/₅ per cent ; about 7 per cent higher than the duty charged on the products called synthetic essential oils. Foreign compounded synthetic essential oils thus enter the country at an advantageous rate of duty, as compared to the raw materials which have been used in their composition and which are required by the indigenous industry.

The number of aromatic chemicals is very large and with continuous research, it is constantly growing. The import control policy permits the import of only a limited number of them falling under specific heads. This seriously reduces the number of aromatic ingredients at the disposal of the Indian perfumer and places him at a disadvantage. Various terpenes and sesquiterpenes, which form important constituents of synthetic perfumes, are not available to him.

NATURAL RESOURCES AND CULTIVATION

India has vast resources in aromatic plants, mostly wild, distributed all over the country. Only a few of them, namely, palmarosa, lemongrass and sandalwood have been systematically exploited, and their essential oils are being exported to the tune of about 5 crores of rupees annually. Some aromatic raw material like coriander, valerian, celery, costus and ginger are exported as such. Vetiver oil of good quality can be produced from the north Indian vetiver.

As the essential oil bearing plants grow in forests, they run a serious danger of hybridization with inferior varieties. The fragrant oils derived from the same species of the uncultivated plant vary widely in odour. In order to obtain products of good uniformity and to improve the quality of the plant material, it is imperative that their systematic cultivation be undertaken. It is suggested that the Government may encourage the cultivators by providing suitable land and financial help.

TESTING AND RESEARCH

The essential oils and aromatic chemicals industry has been suffering from the absence of testing laboratories for standardizing their products. It is gratifying that a beginning has been made by setting up one such laboratory at Cochin.

The perfumery and aromatic chemicals industry is a highly competitive and expanding enterprise requiring constant efforts at innovations. Research is necessary for maintaining a steady pace of progress. It appears that more stress is laid on research in pure chemistry of aromatic products than on the problem directly connected with their industrial utilization. A slight shift of emphasis in this sphere may prove very helpful.

FOREIGN CO-OPERATION

Of the three branches of perfumery, namely, aromatic chemicals, natural essential oils and synthetic essential oils, the aromatic chemicals industry is almost undeveloped in the country. The co-operation of foreign industrial houses in this field may be of benefit to the country and should be welcome. In case of natural essential oils, foreign co-operation in plantation and distillation of materials such as geranium and rose may prove advantageous. The sphere of synthetic essential oils, flavouring essences and perfume specialities should, however, be reserved for indigenous enterprise.

Blending of Popular Perfumes

SHRI CHAND JAIN
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Blending of rose, jasmine, narcissus and *amla* perfumes is discussed. Relevant formulae are given.

The most popular perfumes in India are rose, jasmine, narcissus and *amla*. The first three are obtained from flowers, while the last one is prepared by mixing a number of aromatics, *amla* (myrobalans) having no perfume of its own. Certain types of perfumes of the Chypre-Fougere tone have come to be recognized as *amla* perfumes in the market.

ROSE (*Gulab*)

The real rose odour is unique and represents a type which is undefinable, incomparable and at present inimitable by synthesis. The peculiarity of the rose odour is that no two flowers emit the same fragrance. Different flowers even from the same plant never have exactly the same perfume.

The finest and most powerful form in which the rose perfume can be obtained is the otto, which is a product of distillation. Unfortunately, owing to the loss of one of the constituents, phenylethyl alcohol, a large proportion of which remains dissolved in rose water, the otto does not accurately represent the rose odour which is more exactly represented by the 'absolute' obtained by means of volatile solvents.

With a view to synthesize rose perfume from other aromatics attempts have been made to analyse natural rose otto and the presence of the following constituents in commercial samples have been recorded by different chemists:

Citronellol (principal constituent), 24 to 64 ; total alcohols as geraniol, 63 to 84 ; nerol, up to 10 ; phenylethyl alcohol, 1 ; esters of geraniol, 3.5 ; eugenol, 1 ; and stearoptene, 7 to 25 per cent. Traces of linalol, farnesol, citral and nonyl aldehyde have also been found. The analytical results are in no way complete, but they do indicate the general directions for blending.

A representative formula for rose perfume prepared by the author contains: rhodinol, 5 ; geraniol, 30 ; citronellol, 10 ; phenylethyl alcohol, 15 ; phenylethyl acetate, 5 ; phenyl acetic acid, 1 ; cinnamic alcohol, 2 ; benzophenone, 5 ; Ald. C. 11 (10 per cent), 1 ; oil Bois-de-rose, 4 ; geranyl acetate, 4 ; diphenyl oxide, 4 ; and terpineol, 14 parts with lemon oil, 0.1 part.

Rhodinol is considered the finest base for rose oils ; geraniol from palmarosa oil, nerol, dimethyl octanol and citronellol are good ; and terpeneless oil of rose geranium (French) is also very valuable. Phenylethyl alcohol is indispensable in compounding. Various esters of geraniol are excellent modifiers while rhodinyl acetate and phenylethyl propionate have definite red rose odours. Remarkable freshness is obtained by the addition of such bodies as hydroxy citronellol, α-ionone, and terpineol. Traces of lemon oil are extremely helpful. Fixation can be achieved by the use of musk ketone, patchouli oil, sandal, '*ruh khus*', benzyl isoeugenol, tolu and resinoid benzoin.

JASMINE (*Chameli*)

Many flowers, especially *juhi, bela, chameli, mallika* and *malati* possess the jasmine note. Like rose, the odour of jasmine is unique and cannot be exactly imitated. The discovery of the chemical structure of the jasmine ketone has brought the approximation of the natural odour by synthesis a step nearer.

Chemical examination of the natural jasmine absolute has indicated the presence of benzyl acetate, benzyl alcohol, linalool, linalyl acetate, methyl anthranilate, jasmone, indole and phenylethyl alcohol. Traces of *p*-cresol geraniol, geranyl acetate, geranyl butyrate, benzyl butyrate, nerol, and terpineol have also been reported to be present.

A representative formula for compounding jasmine perfume contains: benzyl acetate, 48.0 ; benzyl alcohol, 10.0 ; benzyl propionate, 2.0 ; linalool, 15.0 ; methyl anthranilate, 1.5 ; linalyl acetate, 7.5 ; indole 100 per cent, 0.5 ; *p*-cresol phenyl acetate, 0.5 ; geraniol, 2.5 ; geranyl acetate, 0.5 ; terpineol, 1.0 ; ylang ylang oil, 3.0 ; benzoin resinoid, 4.0 ; and α-amyl cinnamic aldehyde, 4.0 per cent.

The basic constituent of all synthetic jasmine oils is benzyl acetate, whose odour bears a distinct resemblance to that of the flower. Benzyl alcohol, benzyl propionate and phenylethyl alcohol are useful, while linalool is indispensable. Linalyl acetate acts as a modifier and a sweetening agent. Methyl anthranilate enters the composition of most synthetic jasmines but much better results can be obtained with terpeneless French petitgrain oil. The quantity of indole must not exceed 1 per cent as this substance produces a deep red colour on standing. *p*-Cresol phenyl acetate, geraniol, and geranyl acetate greatly improve the product. In order to obtain a floral note, the essential oil of ylang ylang should be added. Resinoid benzoin helps to maintain the jasmine note and acts as a good fixative. α-Amyl cinnamic aldehyde, when added to a jasmine compound, requires about a month to thoroughly blend ; the quantity should not exceed 3 to 4 per cent. It is a remarkable fixative.

NARCISSUS

The narcissus is a flower with a beautiful perfume, and perfumes described as 'narcisse' are quite popular. A small quantity of the natural perfume is manufactured in the form of *attar*, pomade, concrete or absolute, but the greater part of the so called 'narcisse' perfume is either entirely artificial or contains only a very small amount of the natural product. The odour of narcissus flower is described as resembling that of the violet with the sweetness of the lily. A suggestion of the hyacinth is also usually noticeable.

Chemical examination of natural narcissus absolute indicated the presence of methyl benzoate, benzyl benzoate, indole, methyl anthranilate, esters of cinnamic acid, particularly the methyl ester, and probably linalool. Benzyl acetate, *p*-cresol or derivatives and phenylethyl esters are also suspected to be present.

The blending formulae for 'narcisse' perfume, to be successful, must be based on a natural floral extract or a mixture of extracts chosen from those of narcissus, hyacinth, jonquille, jasmine and tuberose.

The following is a representative formula for 'narcisse' perfume: *p*-Cresol acetate, 2.0 ; *p*-cresol phenyl acetate, 6.0 ; ethylphenyl acetate, 4.0 ; heliotropine, 2.0 ; terpineol, 30.0 ; bromostyrol, 1.5 ; phenyl acetaldehyde 100 per cent, 4.0 ; linalool (pure), 4.5 ; ylang ylang oil, 1.5 ; petitgrain oil, 4.0 ; methyl naphthyl ketone, 2.5 ; indole 10 per cent, 2.0 ; benzoin R, 4.0 ; aldehyde C_{12} 10 per cent, 2.0 ; benzyl acetate, 16.0 ; isoeugenol, 4.0 ; and benzyl benzoate, 10.0 per cent.

The basic constituent of all synthetic narcissus perfumes is *p*-cresol phenyl acetate which bears an odour distinctly resembling that of the flower ; *p*-cresol acetate and *p*-cresol isobutyrate may also be used. Terpineol, isoeugenol, phenylacetic aldehyde, linalool and bromostyrol are good blenders. Phenylethyl alcohol, ionone, methyl anthranilate, ethylphenyl acetate and French petitgrain are often employed as modifiers. The addition of traces of indole supported by benzyl acetate and heliotropine improves the result. In order to obtain the floral note natural absolutes of narcissus, jasmine, tuberose, rose and nerioli must be added. Alternatively, a slight floral effect can be produced with ylang ylang oil. Fixation is completed with the help of benzoin, methyl naphthyl ketone, acetyl isoeugenol, indole or artificial musk.

AMLA

A representative formula for compounding *amla* perfume is given below: Oakmoss, 5 ; patchouli oil, 5 ; sandal oil, 15 ; vetiver oil, 5 ; labdanum, 5 ; musk xylol, 5 ; musk ambrette, 2 ; musk ketone, 4 ; *iso*butyl salicylate, 5 ; hydroxy citronellol, 5 ; bergamot oil, 10 ; safrole, 1 ; petitgrain oil, 4 ; aldehyde C_{12} 10 per cent, 2 ; castoreum absolute, 1 ; vanillin, 2 ; coumarin, 2 ; terpineol, 12 ; jasmine synthetic, 5 ; and rose synthetic, 5 per cent.

Oakmoss, in the form of green or decolorized absolute, is usually employed as one of the basic constituents, although an alcoholic tincture of 10 per cent powdered oakmoss is sometimes preferred. Other basic substances are either the oils or resinoids of patchouli, sandalwood or vetiver. Ionones, Bois-de-rose oil, isoeugenol, *iso*butyl salicylate and cinnamyl esters help to develop the perfume. Flowery notes are obtained with jasmine and rose, the rose generally predominating. Liberal additions of citrus oils give the product sweetness and lift. The fixation is helped by the addition of artificial musks, coumarin, vanillin, dimethyl hydroquinone, styrax labdanum and castoreum.

It is essential to allow a maturing period of at least one month before a blended perfume can be marketed, though a period of two to three months is most desirable. The process of maturing and rounding off can be hastened by the addition of traces of clarysage oil.

Manufacture of Concretes and Absolutes from Indian Flowers

C. GOLDSTEIN
The Tata Oil Mills Co. Ltd., Bombay

The Indian flora abounds in fragrant flowers, but due to lack of scientific investigations natural flower-extracts are not produced on a commercial scale in India. In the absence of their indigenous production the perfume compounding industry cannot be established as an independent entity in this country.

The paper refers to the antiquated extraction methods of indigenous perfumers and gives a list of promising flowers suitable for experimental work. It makes mention of the investigations conducted by the *Tata Oil Mills Co. Ltd.* on the preparation of jasmine concretes and absolutes and reports that the process of light solvent extraction may be applicable under Indian conditions.

The Indian flora abounds in fragrant flowers growing mostly in a wild state. There appears to be a large scope for investigation and development in winning the delicate perfumes from these flowers. Indian manufacturers of flower extracts have so far kept aloof from modern techniques of manufacture. The time has now come to investigate new possibilities, failing which, the perfume industry will be doomed to failure.

Flower extracts are prepared at present in India, particularly in Kanauj, by the following methods:

1. A variation of the distillation process which yields a mild solution of the flower perfume in sandalwood oil. This extract is called an *attar*. The flowers are immersed in water in a copper vessel connected to a receiver plunged in cold water. Sandalwood oil is placed in the receiver. The copper vessel is heated and the mixed vapours of water and perfume are condensed in the sandalwood oil. The perfume dissolved in the oil is separated from water which is reused in the process. Strangely enough such delicate flowers as those belonging to the jasmine family are treated by this process. Roses and *kewda* are similarly extracted.

2. By the digestion of the flowers on the layers of sesame seeds and their subsequent expression. A perfumed oil is obtained and is marketed as hair oil. This is an antiquated process, being the fore-runner of the modern methods used in France. This method yields only solutions of flower perfumes, and not concretes and absolutes.

A concrete is the total extract from the flower having a solid or semi-solid consistency. In addition to perfume, it contains natural waxes, and colouring and resinous constituents.

An absolute is the quintessence of the flower odour. It is prepared from the concrete by extraction with ethyl alcohol. This process is conducted in a suitable apparatus provided with efficient stirring devices. The alcoholic extract, also called 'Lavage' in France is cooled to about – 10°C. and, as far as possible, filtered at the same temperature. This eliminates waxes and resinous matter. The alcohol when distilled off under vacuum leaves behind the flower absolute.

Colourless absolutes are in great demand, and several methods of fractional distillation under almost absolute vacuum have been perfected to yield this type of products.

The success of the French Perfumery Industry in the manufacture of *'Extraits'*, the so called handkerchief perfumes, is due, to a large extent, to the use of the *'Lavages'* as solvents for the finished perfumes. *Jasminum grandiflorum 'Lavages'* are mostly being used.

In order to reproduce the odour of the flower to the best possible perfection, extraction experiments with liquid inert gases at temperatures well below 0°C. have been conducted by the French industry and some of their results have already found commercial application.

The manufacture of concretes and absolutes in India on a commercial scale has not been attempted. The *Tata Oil Mills Co. Ltd.* started experiments on these products in 1942. The flowers treated were varieties of jasmine. The enfleurage process was first tried. It consists in winning the perfume of the flowers in a suitably refined solid animal fat and its subsequent recovery therefrom. Due to climatic conditions and religious prejudices this process does not appear to be suitable for application in India. The work was continued in 1946 on behalf of the Council of Scientific & Industrial Research. Chemical investigation of the flowers of eight varieties of jasmine was conducted by the *Tata Oil Mills Co. Ltd.*

In 1951 the Company decided to carry out extraction experiments with light solvents, a method which appears to be the best suited for this country. Petroleum ether of special grade was used. *Jasminum grandiflorum* (*chameli*) was selected as the flower for the experiment and with a view to collecting economic data on the yield of flowers, scientific cultivation was started on one acre of land at the National Chemical Laboratory farm in Poona. When the flowers were available for extraction, jasmine concrete was prepared for the first time in August 1954 at the National Chemical Laboratory. The yield of concrete was nearly 0.4 per cent.

Jasmine flowers are not the only ones which are likely to offer scope for development. India is well suited by the diversity of its climates and soils for the cultivation of a number of scented flowers finding application in the perfumery industry. By means of petroleum ether or other solvents, the manufacture of concretes and absolutes from the following flowers may be attempted:

The Rose, of which two varieties, *Rosa damascena* Mill. and *Rosa* *centifolia* Linn. are commercially grown at present ; Tuberose, *Polianthes tuberosa* Linn. (Amaryllidaceae) ; Narcissus and jonquill (Amaryllidaceae) ; Hyacinth, *Hyacinthus orientalis* Linn. (Liliaceae) ; Marigold, *Tagetes erecta* Linn. (Compositae) ; Violet, *Viola odorata* Linn. (Violaceae) and Neroli, *Citrus aurantium* Linn.

From the numerous flowers growing wild in India, a few have been selected and listed below. Some of these are already cultivated in gardens. Although perfume extracts from some of them have been investigated by research workers in India, the quantities produced are insufficient to enable perfumers to study their olfactive value.

Acacia farnesiana Willd. (*Divana babul-kikar*), *Acacia arabica* Willd. (*Babul*), *Alangium salviifolium* Wang. (*Akola*), *Brunfelsia americana* Linn. *Caesalpinia coriaria* Willd. (*Libi-dibi*), *Cassia fistula* Linn. (*Amaltash*), *Crotalaria burhia* Buch.-Ham. (*Booi*), *Cestrum nocturnum* Linn. (*Raat-ki-rani*), *Daphne cannabina* Wall. (*Mahadev-ka-phul*), *Ervatamia divaricata* Linn. Alston (*Chandni*), *Lawsonia inermis* Linn. (*Hena mehdi*), *Mesua ferrea* Linn. (*Nag-champa*), *Michelia champaca* Linn. (*Champa*), *Mimusops elangi* Linn. (*Moulsari*), *Murraya paniculata* Jack (*Bisar*), *Nyctanthes arbortristis* Linn. (*Parejatak ; Harsinghar*), *Ochrocarpus longifolius* Benth. and Hook. f. (*Suringi*), *Pandanus odoratissimus* Roxb. (*Kewda*), *Plumeria acuminata* Ait. (*Son Champa*) and *Ventilago madraspatana* Gaertn. (*Bika-Kalibel*).

Concretes and absolutes are concentrated products, and small quantities are sufficient to give body to a perfume. With the advent of aromatic synthetic products, it was felt that the use of concretes and absolutes would suffer a setback. To the contrary, manufacture of concretes and absolutes has considerably increased. Synthetic aromatic chemicals and isolates from essential oils improve considerably in their odour value by the admixture of small quantities of concretes and absolutes. This change is mainly due to minute traces of imponderables in the natural products, which somehow seem to impart exclusiveness to the chemicals and isolates.

Concretes and absolutes are relatively costly products. The price of concretes from jasmine and rose fluctuates around Rs. 1,000 per lb. and that of the corresponding absolutes being double.

I hope that in the interest of the perfume industry, businessmen will undertake the manufacture of concretes and absolutes from flowers in India. For this development it will be necessary to plan with the active support of the Government, agriculturists and co-operative societies. The latter would play an important part in the production of the raw material and its collection.

Production of Jasmine Absolute in India

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The Indian jasmines, on enfleurage, give essential oils which compare favourably with the French oils in 'scent factor' and yield. In order to make the production of jasmine absolute commercially feasible the cost of raising flowers should be reduced by resorting to scientific farming. Recovery of solvents and marketing of by-products will also reduce the cost.

Jasmine absolute is the basic material needed for the production of quality perfumes. Though most of the scented jasmines are indigenous, jasmine absolute is not produced in India. This product is mainly extracted in France, where a big industry is located round about Grasse.

In India the chief centres of jasmine cultivation are Ghazipur, Kanauj, and Jaunpur in Uttar Pradesh, where two types of jasmine products, the hair oil and the *attar*, are prepared. The first product is prepared by impregnating the seeds of *til* (*Sesamum indicum*) with the scent of the flowers and then extracting the oil in a country *ghani* or mill, while the second is obtained by distilling the flowers in earthen vessels and absorbing the aroma in sandalwood oil.

In order to explore the possibilities of producing jasmine absolute in India, the Essential Oils Research Committee of the Council of Scientific & Industrial Research sponsored two research schemes, one dealing with the problems of jasmine agronomy and the other with those of extraction, under the guidance of one of the authors (S. N. Bal). The following observations based on the data collected during the course of these studies throw light on some aspects of these problems and may prove helpful in further investigations.

AGRONOMY

Selection of species—Among the scented jasmines of India, the most valuable species are: *J. grandiflorum* Linn. (*Chameli*), *J. auriculatum* Vahl. (*Juhi*), *J. sambac* Ait. (*Bela*), *J. angustifolium* Vahl. (*Mallika*), *J. officinale* Linn. (*Champa*), *J. humile* Linn. (*Malati*) and *J. pubescens* Willd. (*Kunda*). From experiments it has been found that *J. auriculatum* has a scent factor superior to those of other cultivated jasmines; and the highest percentage of the absolute is obtainable from *J. grandiflorum* cultivated in Uttar Pradesh. In their scent factors and the amounts of quintessence extractable from them, these flowers compare favourably with French flowers; and *J. grandiflorum* can be safely recommended on these bases.

Yield per acre—There are numerous strains of each of the cultivated varieties of jasmines, varying in the productivity, size, showiness and scent-factor of their flowers. To ascertain the most suitable variety for the purpose of cultivation a systematic survey of their yields may be helpful. In France, about 43 maunds of flowers are produced per acre annually whereas in India the average yield per acre per annum of *J. grandiflorum* is from 7 to 8 maunds. Appreciable improvement can be achieved by following improved modern techniques of cultivation.

Soil—Jasmine plants require moderately dry land with irrigation facilities and plenty of sunshine. The soil should be light, friable, well drained and free from gravels. Frost and flood water are detrimental to the plants. The land should be prepared as in the case of cereals. The great alluvial plain of the Ganges from Uttar Pradesh to the Santal Parganas may be considered quite suitable for jasmine cultivation.

Irrigation—Irrigation water should not be sprinkled over jasmine plants but should be supplied through channels. It has been noted that by regulating the water supply, the production of vegetative and floral buds of jasmine can be increased ; and even the opening of the floral buds can be controlled to some extent. In order to study this phenomenon in detail, investigations on the physiology of the plants during floral development may be undertaken.

Manure—Jasmines respond quickly to nutritional stimuli. In India stable manure is generally applied. But in France this practice has long been discontinued and better results are being obtained with ammonium sulphate given with irrigation water.

Grafting—It has been observed in France that grafted plants—*J. grandiflorum* on the root stalk of *J. officinale*—are hardier and more productive. This experience may be of advantage to the cultivators in India.

Pruning—It has been noted that vegetative growth is inversely proportional to the production of flowers. In India about 4,000 to 4,500 plants are put per acre and allowed to grow rather freely. In France, 20,000 or more plants are grown per acre and made into thickset dwarf bushes by skilful pruning.

Harvesting—The quality and quantity of flowers differ considerably according to the season and the time of collection. The most suitable time for collection must be determined for each species of jasmine and the length of the harvesting season ascertained. As wet flowers become unsuitable for extraction, collection becomes a problem during rains. Harvesting, however, can not be postponed as it adversely affects further blooming.

Isolation of Essential Oil

Extraction—Jasmine absolute can best be prepared by enfleurage. As the flowers have very little essential oil at the initial stages and produce it continuously during their life, very little essential oil is obtained by volatile solvent extraction or by other methods which at once destroy the flowers. During enfleurage, the fugitive aroma is captured from the harvested living flowers by suitable absorbent bases.

Enfleurage—For enfleurage wooden frames called "chassis", 5 cm. high and 50 to 80 cm. square, supporting a glass plate in the middle are used. The chassis are placed one above the other. On either side of the glass plate a thin layer of absorbent fat (pomade) is evenly spread, and the flowers, their calyces removed, are spread over the layer with their petals resting on the fat. The spent flowers are daily removed carefully avoiding loss of pomade and the chassis is turned up to expose fresh surface. When the pomade is saturated with the essential oil it is extracted with alcohol ; the little amount of fat and floral waxes which dissolve in alcohol are removed by filtration below 0°C. The alcohol is distilled off under low pressure while the jasmine concrete is left behind. To obtain the absolute, the concrete is further purified by distillation *in vacuo*. The animal fat which is required in the process is freshly collected from slaughtered cattle, washed, ground, rectified and preservatives added to it. After one enfleurage the fat becomes unusable and is sold to soap and cosmetic industries.

The enfleurage process using animal fat gave satisfactory results ; but as a little amount of the absorbent fat remains with the concrete and there is a social aversion to the use of animal fat in India a search for alternative materials was made. Hydrogenated oils and paraffins were found to be unsuitable. The problem may be solved by using pads soaked in vegetable oil in place of pomade. In this case the glass bottom of the chassis may be replaced by a fine wire net. Sesame (*til*) oil or *ben* oil* may prove useful. 'Ben' and olive oils were formerly used in France but due to economic reasons their use was discontinued. However, these vegetable oils have some advantage over animal fats in that they are easier to rectify and have better keeping quality.

Volatile solvent method—The spent flowers discarded after enfleurage still carry some essential oil and some impregnated pomade adhering to them. The essential oil present in them can be recovered by dissolving them in a suitable volatile solvent like industrial alcohol or low-boiling petroleum ether.

Recovery of materials—Excepting a little amount of volatile solvents that is lost, all the by-products are recovered and used in various ways. The pomade or the absorbing base oils which still carry some aroma after extraction may be marketed to soap and cosmetics industries. The floral waxes which separate at the time of preparing concretes or absolutes form a good base material for many cosmetics. The spent flowers can be used as animal feed or fertilizers.

Trade notes—India can produce economically, as good, if not better, jasmine absolute as the French preparation. Jasmine absolute is a costly material. About 85 per cent of the cost is due to labour charges. As a large labour force is required for the production of jasmine absolute, the establishment of this industry may help to solve, to some extent, the unemployment problem of the country.

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*Ben oil can be produced from the seeds of *Moringa oleifera* Lam. (*M. pterygosperma* Gaerten.), which is plentifully found in India. The seeds contain about 36 per cent oil. which can be obtained by simple cold expression. It is not at present produced in India. Besides its use in enfleurage, the oil can be used for various other purposes.

Manufacture of Ionone and Other Aromatic Chemicals in India

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Ionone manufactured from indigenous lemongrass oil at the *Hindustan Fine Chemicals Co.* compares favourably with ionone of foreign manufacture. Its method of manufacture is described. It is suggested that the manufacture of ionone will be feasible in India if the manufacture of some other aromatic chemicals is undertaken simultaneously and some kind of protection is afforded against foreign competition. An estimate of capital required and cost of production at the rate of production of 300 lb. per month is given.

India is the world's leading producer of lemongrass oil. At present most of this oil is exported mainly to America. Lemongrass oil and other aromatic oils produced in the country contain constituents which can be processed into valuable chemicals used in the perfumery and other industries. The author had been producing at the *Hindustan Fine Chemicals Co.*, Bangalore, ionone and other aromatic chemicals from indigenous raw materials on a commercial scale for a number of years. The process used for the manufacture of ionone from lemongrass oil is described below.

Lemongrass oil contains 75 per cent citral, which, on condensation with acetone under suitable conditions, gives pseudo-ionone. Pseudo-ionone on treatment with suitable cyclising agents, such as sulphuric acid and phosphoric acid, gives 100 per cent ionone which is a mixture of α- and β-ionones in different proportions, depending on the method of manufacture.

PROCESS OF MANUFACTURE

Lemongrass oil is mixed with acetone and caustic soda solution and stirred at 60°C. for about 3 hr. The aqueous layer containing excess of acetone, caustic soda, etc. is removed. The other layer containing pseudo-ionone is washed with water and dissolved in a solvent like benzene or petroleum. This solution is treated at a low temperature with 60 per cent sulphuric acid and the resulting 100 per cent ionone purified by fractional distillation at or below 10 mm. pressure. The yield of ionone and its quality depend on the temperature and time taken for condensation, cyclisation and other conditions which should be very carefully controlled. The yield is 40 per cent on the weight of lemongrass oil.

COST OF MANUFACTURE OF IONONE 100% (300 LB. PER MONTH)

	Rs.	As.	P.
A. CAPITAL REQUIRED			
1. Plant and other capital equipment	30,000	0	0
2. Working capital	30,000	0	0
	60,000	0	0
B. RAW MATERIALS			
1. Lemongrass oil—750 lb. @ Rs. 5/- per lb.	3,750	0	0
2. Acetone—1018 lb. @ As. -/14/- per lb.	891	0	0
3. Solvent oil or petrol—804 lb. @ Rs. 4/8/- per gal.	457	0	0
4. Caustic soda—32 lb. @ As. -/6/6 per lb.	13	0	0
5. Sulphuric acid commercial—300 lb. @ As. -/3/9 per lb.	70	0	0
6. Salt—5 bags @ Rs. 8/- per bag	40	0	0
7. Ice—6500 lb. @ 9 pies per lb.	305	0	0
	5.526	0	0
Less lower fractions and residues—450 lb. @ Rs. 2/- per lb.	900	0	0
	4,626	0	0
Cost per lb. of ionone	15	7	0
C. OTHER EXPENSES			
1. Interest on capital (Rs. 60,000) @ 4%	200	0	0
2. Rent	100	0	0
3. Electric power	50	0	0
4. Water	50	0	0
5. Technical staff	200	0	0
6. Labourers, clerical staff, etc.	300	0	0
7. Other expenses	100	0	0
Total	1,000	0	0
COST PER LB.			
Cost of raw materials per lb.	15	7	0
Labour, overhead, interest and other expenses @ Rs. 1000 per month	3	10	8
	19	1	8
Cost of material per lb.	19	1	8
Bottle, packing, freight, etc.	1	7	4
Total cost per lb.	20	9	0

The 100 per cent ionone manufactured at the *Hindustan Fine Chemicals Co.*, was tested by independent analysts in England and found to be equal or superior in quality to the best foreign makes.

Some authors recommend that pure citral should be used instead of lemongrass oil, but this has been found to be unnecessary. It is also unnecessary to isolate pseudo-ionone and purify it before cyclisation. All the impurities can be removed in the final vacuum distillation, if an efficient fractionating column is used and the temperature carefully controlled.

α- and β-ionones can be separated for special purposes by making use of the difference in properties of their sodium bisulphite compounds. Recently β-ionone has assumed great commercial importance as the starting material for the manufacture of synthetic vitamin A.

All the equipment used for the manufacture, with the exception of a high vacuum pump, was manufactured locally. The condensation of citral into pseudo-ionone was carried out in a jacketed mild steel vessel fitted with a mechanical stirrer and heated by electricity. The cyclisation with sulphuric acid was carried out in lead-lined vessels fitted with stirrers and cooling done by means of a freezing mixture. The purification of ionone by fractional distillation was carried out in an electrically heated still (monel metal or stainless steel) fitted with an oil jacket, an efficient fractionating column filled with Lessing rings and a dial thermometer. A small plant costing Rs. 25,000 to Rs. 30,000 is capable of producing 300 to 400 lb. of ionone per month. As all the raw materials required are produced in India and most of the equipment can be fabricated locally, the manufacture of ionone appears to be economically feasible, provided some kind of protection is given to the industry against foreign competition and the manufacture of other aromatic and fine chemicals is undertaken simultaneously.

Standard Methods of Test for Essential Oils

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The standard methods of test for essential oils prescribed by various countries and those proposed by the International Organization for Standardization (ISO) are discussed and compared. The work done by the Indian Standards Institution (ISI) in this field is also reviewed.

Bulk for bulk, essential oils and materials which contain them fetch higher prices than other naturally occurring materials. Their quality and characteristics are, however, subject to appreciable changes due to their origin and methods of production. It is, therefore, imperative to define their quality by means of measurable physical and chemical characteristics.

Their chief value is in their odour, but odour is not capable of precise measurement.

Odour evaluation is a kind of fine art and is too subjective to serve as a test of quality. Composition would provide a useful guide but the composition of essential oils, as of most other naturally occurring organic materials, is very difficult to determine or assay and is time-consuming. It becomes necessary, then, to fix limits of physical and chemical characteristics for essential oils which are held to be of acceptable odour value and to test different samples to these limits of requirements to establish their quality.

Data of physical and chemical characteristics of essential oils obtained from different sources during different seasons and by different processes, have been accumulating for about 90 years. Masterly treatises, well known to essential oil chemists, have held sway one after the other, but the oldest of these are still valuable source of information. All of them have contributed to further research and discovery of fresh materials. The data given in these publications are of great value in fixing specifications of essential oils, but the techniques of analysis require standardization. Standards on comprehensive methods of test had not been issued by any national standardization authority till recently.

The International Organization for Standardization (ISO) had established a Technical Committee known as ISO/TC 54, but its work was dormant for want of an active secretariat.

In the meantime, the Indian Standards Institution set up under the Chemical Division Council a Sectional Committee for Essential Oils, briefly designated and spoken of as CDC 11. It may be mentioned in passing that the Secretariat of ISO/TC 54 fell vacant and that India just missed the chance of taking up the Secretariat because Portugal made an offer before India. Before taking up material specifications, CDC 11 set up a sub-committee for Methods of Test with Dr. Srikrishna as convener. Working documents were prepared by Dr. Srikrishna and these were put into shape as Draft Indian Standard Methods of Test for Essential Oils. They were circulated in India and abroad for a period of three months to all those who were interested with a view to eliciting comments for their improvement. The draft was finalized by CDC 11 in August 1951 and published as Indian Standard Methods of Test for Essential Oils (IS: 326-1952). They have proved to be of considerable assistance in the subsequent issue of Indian Standards on a number of essential oils.

The British Standards Institution also issued in 1953 a comprehensive British Standard (B.S. 2073: 1953). The ISO/TC 54 is taking up these methods one by one. In this connection, the member countries, who participate actively in the work of ISO/TC 54, issue their national documents which give an instructive insight into the uniformity which prevails in an apparent international diversity. It is of interest to examine the standards on methods of test issued by the various authorities and to assess how far there is agreement between them.

STANDARD METHODS AND RESEARCH METHODS

There is a subtle but important difference between the use of a method for purpose of investigation and research on the one hand and for testing the acceptability in trade of an industrial product. While, for the former, we have to try and reach perfection, for the latter we have also to keep in prominent view the extent to which the method is in vogue, how easy it is to test according to the method, its repeatability by two different persons in the same laboratory and its reproducibility by different persons in different laboratories. In view of these considerations, a standard method of test may not be a perfect method. This may sound like heresy in scientific circles, but when we consider the method in actual practice and examine this statement, its justification becomes more obvious. In the exchange of goods, we have to judge any characteristic by limits. We do not fix an absolute value; we prescribe a range. In fact, absolute value does not exist for an essential oil. To determine a valid range for any characteristic, we have to examine all available data. If, therefore, data have been collected, using largely a fairly commonly accepted method, it follows that the limits derived from such data would be valid only if the particular method is followed. A research worker pins his objective on accuracy and novelty; a standardization technologist is more interested in agreement and ease of operation.

Taking any of the common determinations employed in characterising essential oils, we find that though, in concept, the methods followed by different laboratories and groups are similar, they differ in the matter of detail.

Yet, for standard methods, we must have complete identity. This point will be illustrated later on.

CHARACTERIZATION OF ESSENTIAL OILS

The characteristics, which are of importance in evaluation of essential oils are: (*i*) preliminary examination of colour, clarity, separated water and by-notes which are largely subjective, (*ii*) specific gravity, (*iii*) optical rotation, (*iv*) refractive index, (*v*) solubility in ethyl alcohol of various strengths, (*vi*) acid value, (*vii*) saponification value and ester content, (*viii*) saponification value after acetylation, (*ix*) percentage of alcohols, (*x*) aldehyde and ketone content, (*xi*) phenol content, (*xii*) tests for heavy metals and (*xiii*) presence of adulterants, such as petroleum products, fatty oils and cheaper essential oils like gum spirit of turpentine and cedar-wood oil. Methods of test for these have been prescribed in IS: 326-1952. These methods are of general application but when dealing with a particular oil, it might be necessary to introduce either entirely different methods or some modifications of the standard method. These features are taken care of by issuing, as an appendix to the material standards on essential oils, where necessary, a special method which supersedes the general method.

COMPARATIVE STUDY

The Indian Standard Methods of Test, as contained in IS: 326-1952, may now be taken up for comparison with other recognized methods of test. There may be portions of the Indian Standard requiring revision, but the ISO documents are only at the discussion stage and it will be advisable for India to wait till final international decision is reached before attempting a revision.

Preliminary examination—Precise definition of quality is not possible by preliminary examinaton. It is used by the trade on a conventional basis.

Specific gravity—The Indian Standard prescribes a temperature of 30°/30°C. ; ISO document prescribes 20°/20°C. and the British Standard, while prescribing t°/20°C. gives 't' the value of 30°C. for three oils and 20°C. for others. The ISO and BS conditions are easy to attain in temperate and cold climates but not in the tropics. India had urged that a supplementary temperature of 30°C. should be permitted in tropical countries, but ISO/TC 54 has not accepted our view. The ISI will pursue the matter still more vigorously. As it is not always easy to secure a thermostat set at 30°C. the Indian Standard permits the determination being made at ±3°C. of 30°C. and the value corrected for 30°C. by the use of a correction factor which is given for each oil in the material standard relating to that oil.

Optical rotation and refractive index—There is general agreement on this method of test. The temperature of test is important. Here too the Indian Standard prescribes 30°C. and permits the use of a correction factor. The ISO committee prescribes a temperature of 20°C. and has not yet accepted the suggestion to introduce a supplementary temperature for tropical countries.

Solubility in alcohol—The volume concentrations used in the Indian Standard are 50, 60, 70, 80, 90 and 95 per cent. The ISO document uses these concentrations and interposes concentrations of 55, 65, 75 and 85 per cent also. The British Standard uses 60, 65, 70, 80, 90 and 95. The British Standard and the ISO documents prescribe only one standard of opalescence as obtained by adding 0.5 ml. of 0.1 N silver nitrate solution to 50 ml. of 0.0002 N sodium chloride solution. India prescribes three standards; faintly opalescent, slightly opalescent and distinctly opalescent of which the one characterized as slightly opalescent is the same as prescribed by the British Standard. The Indian Standard goes farther still and prescribes three standards of turbidity by diluting 0.25, 0.5 and 1.0 ml. of 0.1 N sulphuric acid to 50 ml., adding 0.2 ml. of normal barium chloride solution and allowing to settle for 5 minutes. The Indian practice had been commented upon favourably by essential oil technologists.

In the ISO document, solubility is to be tested by adding up to 20 volumes, while in the Indian Standard we add only up to 10 volumes.

Acid value—The basic method is the same everywhere. In India, we use aqueous potassium hydroxide solution and so does the Essential Oil Association of USA, but the British Standard and the ISO document specify the use of alcoholic solution. This enables them to proceed direct to the saponification value.

Saponification value—All the methods agree.

Saponification value after acetylation—The methods followed by different authorities are similar. The quantities of reagents used for acetylation in different methods are given in Table 1.

For the determination of saponification value of the acetylated oil, British Standard and the ISO document prescribe the use of 40 ml. of 0.5 N alcoholic caustic potash solution, while India, E.O.A., France and Netherlands use 25 ml. India in its comments on the ISO document, has stated that the quantity of sodium acetate may be raised to 2 but the use of 20 ml. of acetic anhydride is unnecessary.

In both these operations, India's viewpoint is that for the standard method, the average quantities and not the maximum required should be prescribed. In particular essential oils, where larger quantities are required, the position can be covered by prescribing the recommended quantities in the material standards.

TABLE 1—QUANTITIES OF REAGENTS USED FOR ACETYLATION OF ESSENTIAL OILS IN DIFFERENT METHODS

	IS	BS	EOA	ISO DOCUMENT	FRANCE	NETHERLANDS
Vol. of oil, ml.	10*	10	10	10	10	10
Wt. of anhydrous sodium acetate, g.	1	2	1	2	2	2
Vol. of acetic anhydride, ml.	10	20	10	20	10	10
Duration of heating, hr.	2	2	1	2	1	2

Determination of aldehydes and ketones—All authorities prescribe the hydroxylamine method. France and India give the neutral sulphite method as an additional method. India has also prescribed the sodium bisulphite method which can be used conveniently even in the field. India is pressing for the acceptance of this method particularly for lemongrass oil.

Determination of phenols—All the methods are similar.

Determination of cineole—All the authorities adopt the *o*-cresol method of Cocking. This method gives relatively more accurate results for oils with a high cineole content. In Netherlands, the preparation of a cineole-cresol complex is prescribed and the complex is added in known proportions to boost up the cineole content of oils containing less than 50 per cent cineole. Oils with a known high cineole content may be used for the preparation of the complex. The Indian method prescribes the use of pure, dry *o*-cresol.

General—The Indian Standard prescribes methods of test for detecting the presence, as adulterants, of petroleum products, turpentine and cedar-wood oil. Methods for detecting the presence of fatty oils have also been indicated.

To conclude, India has a good working standard for methods of test of essential oils. In the international field too, there has been a definite quickening in formulating standards for essential oils. Alternative proposals are emanating from other countries, largely under the inspiration of United Kingdom. We must receive them with an open mind and try them out. There are several very important centres of research on essential oils in India. We should ask them to band themselves as a team for investigating the new proposals so that what is good in them may be adopted and what is not acceptable may be proved to be so on the strength of scientific data. India and Arabia have been famous for their perfumes since ancient times. Arabia came into the picture more as a purveyor of perfumes than as a producer. India and the East have been the home of essences and spices. In the regeneration that is now afoot in our country, we have to aim at competitive economy in international trade. For this we need standards on essential oils and, for ensuring quality, we must help in developing not only national but international standard methods of test for essential oils.

Indian Standards for Essential Oils

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The work of the Essential Oils Sectional Committee (CDC 11) of ISI is outlined. The difficulties encountered while formulating standards for palmarosa oil, lemongrass oil, eucalyptus oil, gum spirit of turpentine, vetiver oil, imported essential oils and gum rosin are discussed.

From historical accounts it is evident that the use of some of the essential oils was known in India in Vedic times, well before the dawn of the Christian era. It appears that India produced sufficient amount of essential oils to enable her to export them. The flourishing trade of ancient and mediaeval times, however, receded because of lack of scientific and technological progress. The introduction of cheap aromatic chemicals and synthetic perfumes helped this regression. In recent times, with the realization of the importance of this industry in India's export trade, efforts are being made to place it on a scientific basis right from the cultivation stage to the production of the final product.

The industry, as it exists, is not well organized and consists mainly of small-scale producers, although a few large-scale concerns, like the Mysore Sandalwood Oil Factory, do exist. Production is carried out, as in the case of turpentine industry, chiefly using oil methods. They are often wasteful and sometimes so outmoded as to hinder rather than help the industry.

Need for Standards

It is being realized by Government, as well as by the producers that the industry requires scientific aid for its growth. Among the several steps that are necessary to encourage the growth, undoubtedly standardization is one of the most important. This will help the production of a more uniform product acceptable in the market and prevent wastage of effort and materials. Standardization will stimulate internal trade and will lessen disputes in export business. Besides, it will enable the importers to judge the quality of the materials imported. Standardization caters to the widest sector of producers and consumers, and with the minimum of hardship, will go a long way towards improving the status of the essential oil industry in India.

WORK OF THE INDIAN STANDARDS INSTITUTION (ISI)

With these and similar considerations in view, the ISI set up in September 1948, the Essential Oils Sectional Committee (CDC 11). This Committee has since then prepared standards for a number of essential oils and allied products such as, lemongrass oil, eucalyptus oil, sandalwood oil, citronella oil, palmarosa and gingergrass oils, rectified camphor oil, peppermint oil, ginger oil, vetiver oil, gum spirit of turpentine and gum rosin. In the production of these standards full use has been made of the existing data on the constants, methods of test for essential oils, production methods, and also the views of the producers and consumers. In quite a few cases, it was even necessary to carry out considerable investigational research before arriving at the final requirements.

It may be easier to comprehend the effort that has gone into these standards if we consider the background of the work on the standards and analyse the difficulties which had to be overcome.

As is well known, sandalwood, palmarosa and lemongrass oils figure very prominently in the export trade of the country.

Sandalwood oil—Sandalwood oil manufacture is well established and annual production is nearly 150 tons, most of which is exported. Standardization of this oil involved the least difficulty as reliable data were readily available.

Palmarosa oil—Though India is one of the principal producers and exporters of palmarosa oil in the world, attempts to evolve a scientifically correct and acceptable standard for the pure grade oil have been thwarted because of the widely divergent values reported in literature for pure palmarosa oil due to different proportions of flower heads to stalks of the grass used by various distillers. Another reason is the adulteration of the oil, either through admixture of the allied gingergrass in the field and at the time of distillation or, later, by the mixing of gingergrass and palmarosa oils in the trade. Therefore, it involved considerable care and scrutiny of data available on both gingergrass and palmarosa oils before a standard could be laid down for the content of geraniol, which is the principal constituent of palmarosa oil, ensuring the proper grade of rose-smelling palmarosa oil. The standard, however, excludes a variety of palmarosa oil produced in Hyderabad and Madras with a geraniol content of 60 to 80 per cent. It is hoped that with further research and development, it will be possible to produce an oil with a geraniol content of 90 per cent or to rectify the oil now produced in Hyderabad and Madras to meet the requirements of the Indian Standard.

Lemongrass oil—The Indian lemongrass oil is produced mainly in Travancore-Cochin and is valued in export trade because of its high citral content and good quality. The solubility of the oil in 70 per cent alcohol is also important, as fresh and unadulterated oil is soluble in 70 per cent ethyl alcohol. Most of the Indian lemongrass oil conforms to the solubility requirement but genuine samples are sometimes found to be insoluble in 3 volumes of 70 per cent alcohol. Here also considerable data were available but sifting was necessary before a standard could be evolved.

Eucalyptus oil—Controversy has arisen in the preparation of a number of standards for essential oils. In the case of eucalyptus oil, cineole content was the chief point for consideration. The *minimum* cineole content prescribed in British Pharmacopoeia (BP) is 70 per cent but the Indian Pharmacopoeial List (IP) 1946 lays down the limit at 55 per cent to accommodate the material as distilled in India from Indian eucalyptus trees. As this lower limit of cineole content facilitates the use of the indigenous material in pharmacy, it was adopted for the present, with the hope that a 70 per cent cineole content would be attained in future by suitably modifying the manufacturing process to include a simple rectification of the oil. The standard has, therefore, been kept tentative*.

Gum spirit of turpentine—Another case in point is the standard on gum spirit of turpentine, or the turpentine oil of commerce. India produces more than 2,000 tons of turpentine and exports some of the product. In laying down the standard two grades were recognised: Grade 1 for pharmaceutical use and Grade 2 for use in paint industry ; it is hoped, however, that Grade 1 would be preferred for the manufacture of high class paints. It was also the intention of the Committee to discourage the use of such terms as 'rectified oil of turpentine' or 'double distilled oil of turpentine', as they are meaningless and misleading. Before arriving at the above conclusions it was necessary for the ISI to convene a conference of turpentine oil producers to assess the methods used in production, grades produced, analytical data for each grade, possibilities of better class material being produced and the steps necessary to reach these objectives. It was only at the conclusion of these consultations, that any definite progress could be made. It was revealed in this enquiry that appreciable quantities of low grade turpentine are produced in India, some by the crude old-fashioned methods, some by using outmoded equipment. It is, however, gratifying that the production of gum spirit of turpentine is likely to be placed on a more scientific and progressive basis in the near future in Government owned and/or aided factories as also in private factories. Considerable research was also necessary in formulating requirements, because the composition of Indian turpentine oil is different from that of the oil of foreign origin.

Vetiver oil—An example of a standard catering to a product which, in due course, is capable of much improvement is the standard now under preparation for vetiver or *khus* oil. The grass grows wild in India, and the quality of the oil which is obtained from the wild roots differs from place to place. Consequently, considerable amount of study was necessary before a standard could be proposed. Such investigations were carried out at various places, chiefly at the Forest Research Institute, Dehra Dun. It is now possible to draft a standard on the basis of these investigations for the vetiver or *khus* oil derived from wild grass. The need for scientific plantation on modern lines has been keenly felt and it is hoped that it will receive encouragement. It is gratifying that cultivation of the grass is already being taken up in Travancore-Cochin while the Uttar Pradesh Government are taking steps in the same direction.

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*The Indian Standard on Eucalyptus Oil has since been revised and cineole content raised to 60 per cent in view of this limit being accepted by the Pharmacopoeia of India. Some other requirements have also been revised concurrently.

Imported essential oils—Besides the standards which are based mainly on data collected in India for Indian products, the Essential Oils Sectional Committee has also prepared standards for some essential oils which are often imported into India because the indigenous production is small and is not sufficient to meet the demand. Examples are the standards for cintronella oil, rectified oil of camphor, peppermint oil, geranium oil and ginger oil. The requirements for these materials are based on the accepted overseas standards and it is hoped that these standards would help in the development of indigenous production of these oils.

Gum rosin—The work of drafting a comprehensive standard for rosin (gum rosin) was also entrusted to the Essential Oils Sectional Committee. Although a standard for rosin for use in paints was already published, it was felt that a comprehensive standard for rosin for use in rubber, paper, soap and other industries was desirable. The name gum rosin, like gum spirit of turpentine, was adopted to distinguish the rosin obtained in the distillation of oleo-rosin of *Pinus longifolia* from rosin obtained from other sources. A feature of this standard is that the colour values of the various grades of gum rosin would be expressed in terms of Lovibond units as they are more easily understood than the values expressed in terms of spectral transmission factors. The United States Department of Agriculture and Messrs *Tintometer Ltd.*, England, have extended their co-operation to the ISI in this work, and the ISI would now be able to lodge in the Forest Research Institute a prototype standard set of Lovibond glass cubes for colour comparison of various grades of rosin. Secondary standards made of selected grades of rosin would be matched with the prototype and made available to the public by the Forest Research Institute.

Research and standardization—From the foregoing it is apparent that careful and extensive study had to be undertaken for evaluating the constants of various essential oils produced in India. It is gratifying to record here that the co-operation of the research laboratories in India has been forthcoming readily in this work. The work done at the Forest Research Institute, Dehra Dun, National Chemical Laboratory, Poona, Harcourt Butler Technological Institute, Kanpur and *Tata Oil Mills Co. Ltd.*, Bombay, has made it possible to formulate a number of standards.

Sense of Smell and Molecular Vibrations

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The science of olfactics is not yet developed. The problems facing the study of the sense of smell and the recent trend of thought in the field are outlined. The close relationship between odour and Raman effect is explained and a new approach to the problem through the study of Raman shifts and the infra-red absorption spectra of odorants is suggested. The study of photochemical reaction on odorants and their intramolecular vibration patterns is indicated.

Olfactory nerves are directly connected to the brain and some physiologists contend that the brain began as a smelling organ. It looks probable that in the process of evolution the sense of smell developed first. Observations show that babies up to a fortnight after their birth, respond more readily to odour stimuli than to optical or auditory ones.

Our knowledge of the sense of odour is very meagre. It is not definitely known how the sensation of smell is produced. Nor do we know the agents which produce the characteristic sensations of pleasant or unpleasant odour. Perhaps the sensation of odour is a complex association of sensations. The science of olfactics is yet to be developed. Various factors relevant to it have to be measured and given a physical and mathematical treatment.

The questions which face us in the study of the sense of smell are numerous. What are the properties of a substance which make it odorous? Are they physical or chemical? Which of them are responsible for its various odours? How is the sensory perception effected? Are material particles responsible for the sense of smell or is it induced by some kind of radiation? How is the message carried to the brain? How many fundamental odours are there and how can they be measured or recorded?

Many theories have been advanced on these points. The main issue has been whether the sensation of odour is due to the actual particles of the odorant reaching the sense organ or whether it is due to some kind of ethereal waves. According to the modern trend of thought, when the volatile and lipoid-soluble odorant molecules of a substance come in contact with the olfactory region, they stimulate the receptors through their intramolecular vibrations. It is not the chemical constitution or structure of the molecule that is responsible for a certain type of odour. It seems that the intramolecular frequencies or the rotational or vibrational movements of the nuclei and electrons of the atoms present in the molecule induce the sensation of odour.

Dyson[1] has shown a close relationship between odour and Raman effect. When light of a definite frequency, such as the monochromatic light from a mercury vapour lamp, is made to fall on a body, the scattered light is no more monochromatic. When examined by a spectrometer, the scattered light is found to consist of both shorter and longer wave lengths than that of the original incident light. This change in wave length is the Raman shift. These shifts are independent of the wave length of the incident light, and are expressed as the number of vibrations per centimetre. Dyson[2] has shown that substances which have Raman shifts between 1,400 and 3,500 are odorous provided they are lipoid-soluble and volatile. But the recent findings of Wright[3] indicate that 'any correlation between odours and molecular vibration patterns must be looked for at frequencies below about 700^{-1} cm. corresponding to absorption in the infra-red beyond about 15 microns. A correlation of odour with the pattern of molecular vibrations may therefore be possible if the frequency range below 1000^{-1} cm. is studied. Raman effect is at present the most convenient tool for the purpose'.

If the sensation of odour is produced by the intramolecular vibrations of the odorant molecules and Raman shift is also due to the molecular vibrations, then Raman shift should naturally throw more light on the nature of odours that are caused by these molecular vibrations. Raman shift being an optical effect offers a visible map of the intramolecular vibrations. Any interference with the vibrations of these molecules or any resonance effect should be accompanied by odour alteration-diminution or accentuation.

Certain substances when smelt together do not give the sensation of smell, but individually they are found to be odorous. This phenomenon of cancelling out the odours cannot be ascribed to chemical change, because it is produced when two odorants are smelt separately at the same time by the two nostrils. According to Stoves[4], well-known neutralizing pairs are cedar-wood and rubber, and paraffin and rubber, while 4 g. of iodoform in 200 g. of balsam of Peru yield an almost odourless mixture.

It is found that light changes odour. The incident light energy may be bringing about a change in the rotational movement of the odorant molecule. This may suggest the study of the effect of incident light or other electromagnetic radiations on the odorous character of the molecule. It may be possible to find out the relation between the incident energy and the intramolecular vibrations, the balancing of the energy potential and the resultant odour.

Since all molecules show these intramolecular vibrations they must all be potential odorants. But all substances are not odorous. Therefore, we may have to assume that our sense of smell can perceive odours within a certain range. Dogs, other animals or insects may have different ranges of perceptivity. It is likely that by photochemical reaction on odorants the frequency of intramolecular vibrations is altered and if it falls outside the perceptible range of osmic frequencies the substance is no more odorous to our nose. There is the other possibility that an inodorous substance may become odorous by irradiation when the adjustment of energy potential of the molecule becomes such that the frequencies fall within the osmic frequencies. Much experimental work is necessary to test the validity of this hypothesis.

If we study the intramolecular vibration patterns of certain odorants which produce masking or enhancement effects due to resonance with the help of Raman shifts and absorption spectra particularly with infra-red light of about 15 microns, we may find more closely the causes to which can be attributed the sensation of odour.

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Role of Odour in Mental Dynamics

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The odour of perfumes can influence the region of the brain which is not under conscious control but is linked up with our emotions and moods. Perfumes have therapeutic properties, and are used in various industries to mask unpleasant odour and give a pleasing aroma. The sense of smell is very powerful as a sales force. It is suggested that an organization should be started to promote the cause of the perfumery and aromatics industry through consumer education.

The sense of smell plays an important part in our activities. It is said that the nerves which carry the message of smell are the only ones directly connected with the brain and that the brain began as a smelling organ. In a recent work[1] olfaction is described as a short circuit sense. The memory associations of smell are deep and subtle.

The olfactory sensation is of a more subjective nature. Odours or perfumes can influence the region of the brain which is not under conscious control. This perception affects our psychic life and subtly moulds our moods and dispositions. No man is insensible to the pleasurable effects of a good perfume. The psychological aspect of the perfume with regard to its pleasure-giving quality is called hedonics. The feeling of pleasure derived from the sense of smell depends on the mind of the subject. There are primitive people who use butter as hair oil, and the more rancid the butter, the more they like it. The idea of pleasure derived from odours also changes with times. In the days of Louis XIV, the liking was for spicy, resinous and incense-like aromatics. Nowadays mild floral perfumes and exotic blends are preferred. The recent trend in France is towards the aldehydic top note rather than the floral type.

Psychologists have observed that perfumes have a great influence on the sex impulse. It is said that women are more susceptible to the olfactory stimuli than men. Physicians, particularly neurologists, know well the value of perfumes and their physiological effects; they have both preventive as well as therapeutic properties.

Perfumes are a useful means of protection against disease producing bacteria. The use of eucalyptus oil against the common cold is well known. Citral occurring in lemongrass oil is reported to inhibit tumour growth in cancer. With the aid of aromatic substances diffused in the air conditioned theatres and halls, the chances of people catching cold or influenza can be minimized.

It has been reported that lilac, lavender and heliotrope have favourable effect on the recovery of patients convalescing after operations. Children in hospitals are benefited by the soft and soothing odours of lily of the valley and sweet pea. Sandalwood is said to be beneficial in subduing the nervous tremors accompanying some kinds of fever. In India sandalwood paste with other odorants is applied on the forehead in headache and fever. Lavender water, Eau-de-Cologne and *attar* of roses have been found to be very efficacious in cases of brain fag and headache. The balsamic odours of pine are very soothing in insomnia.

Odours have a great influence on the digestive system. Some pleasant odours stimulate the appetite, while the unpleasant or disturbing ones can impair it. There are certain odours like peppermint which accelerate the process of digestion. In a hot country like India, where the bacterial activity is very rapid and the contents of the intestinal tract are quickly subjected to fermentation producing toxins, the spices are useful in checking the decomposition and neutralizing its products. *Tambul* contains several substances rich in essential oils which contribute to oral hygiene, and also serves as digestive and anthelmintic agent. In disease and under emotional stress, a person gives out characteristic odours which form a subject of study for doctors and psychologists.

A study of the mental reaction to odours would be very fascinating. Why does our mind react in a certain way to an odour? It may be a complex mental process. As this subject is on the borderland of psychology, experiments to study the psycho-galvanic reflex (P.G.R.) with respect to a particular type of odour may be undertaken.

The process of smelling is not always a conscious one. We may be smelling many things which we do not consciously note; but these fleeting odours can exercise a subtle influence on our unconscious makeup. These subtle odours enter the subconscious mind by a short route and influence the thoughts without our being aware of them. This fact can be exploited by the perfumer, particularly in the field of advertisement and sales.

An interesting phenomenon may be mentioned. When people are asked if they like scented commodities, they resent the idea and assert that they prefer the unscented ones ; but actually they unconsciously choose the mildly scented articles. This fact is very important. The prospective buyer does not like to be made conscious that he has been influenced in his choice by the pleasant odour of the material. This is the subtle role of odour in the mental dynamics of human beings.

Perfume adds to the joys of life and can make unpleasant things more tolerable or pleasant. In industry aromatics have been used for this purpose. Many of the products of petroleum industry have unpleasant odour. There are residual unpleasant odours left in the products of textile, leather, paper, rubber and plastic industries. To make the commodities more attractive and to increase their sales appeal, it is necessary to mask the unpleasant odours by some pleasant fragrance. The textiles and laundered clothes would be more acceptable if they have a faint but delightful aroma.

In paper mills, breweries, fruit canning and fish processing plants also, deodorants and aromatics are used to cover the unpleasant odour and to impart a pleasant note. The possibilities of application of aromatics in industries and other fields of human activities appear to be unlimited.

An organization should be set up to promote the cause of the perfumery and essential oils industry through consumer education and to study applications and uses of aromatics. This organization can also undertake market research and surveys, and maintain a library of odours.

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Essential Oil Resources of Assam

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The production of essential oils from *Pinus khasya, Aquilaria agallocha, Ocimum kilimandscharium* **and** *Santalum album* **in Assam has been discussed.**

Assam with its wide range of climatic zones from the semitropical to the almost Alpine, possesses a great variety of indigenous flora, yielding both essential and fixed oils. Some of the plants are being commercially utilized and some are too scattered to be of use for economic exploitation. Attempts are also being made to cultivate some of the exotic species yielding these oils. An account of the plants being exploited at present and those that are capable of exploitation is given below.

Turpentine oil from *Pinus khasya* (khasi pine)

Khasi pine (*P. khasya*) is found in Assam in the Khasi and Jaintia Hills, wherefrom it derives its name, as well as in the Abor and Mishmi Hills in the North East Frontier Agencies, in the Balipara Frontier Tracts, Naga Hills, Lushai Hills and in Manipur above an altitude of about 3,000 ft. The acreage under this pine is not correctly known but it is available to quite a large extent especially in the Frontier areas. In the past, little effort was made to utilize this source of turpentine and rosin. In 1953 a pilot plant for distilling one ton of pine resin per day was set up in the Khasi and Jaintia Hills in Shillong. During the first year of its working, it produced about 4,000 gallons of turpentine oil and 2,000 maunds of rosin. The peculiar weather conditions in these Hills have hampered the extraction of the pine resin from the forests. The yield of resin per channel could not be increased much above 2.0 lb. per season from October/November to June/July. The season in Assam is different from that of Uttar Pradesh and the Punjab where pine resin is tapped from March to November. Cold weather tapping has been resorted to in the Khasi and Jaintia Hills in order to avoid the heavy monsoon rains averaging 120 in. from June-July to August-September. The low yield of resin, to some extent, is also due to the inexperience of the tappers. During the two seasons starting from 1952-53, tapping has been confined to the

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Government forests extending over an area of about 3,000 acres in and around Shillong. Attempts are being made to induce private pine growers in the district to take up resin collection as part time work in their own forests. Private growers own most of the pine areas of the Khasi and Jaintia Hills. With their coming in, resin tapping may prove a success and eventually the industry may spread over to the other pine areas of the State including Manipur and the North East Frontier Agencies.

The resin obtained from the Khasi and Jaintia Hills yields 19 to 20 per cent turpentine oil and 65 to 70 per cent rosin. The turpentine oil was examined at the Forest Research Institute, Dehra Dun, and found to be of a very good quality. It is a colourless mobile liquid with a fine piny smell free from tackiness and unlike turpentine oil from *P. longifolia* contains a higher percentage of low boiling terpenes. Different samples examined showed sp. gr.$^{30°}$, 0.8648-0.8712 ; $n^{30°}$, 1.4632-1.4645 and $[\alpha]_D^{30°}$, $-3.83°$ to $-4.12°$.

Two fractions distilling between 155° and 156.5°C. were totally converted into the solid crystalline hydrochloride derivative, which on recrystallization melted at 131°C. ; the melting point of the corresponding derivative of α-pinene being 132°C.

From the data given below (Table 1) it appears that the two oils obtained from *P. khasya* contain α-pinene to the extent of 77 per cent and 71 per cent respectively, and may prove an economical source for the production of important synthetics like camphor and terpineol.

Subsequent examination of samples of the turpentine oil produced at the Assam Government Rosin and Turpentine Unit at Shillong and by the National Chemical Laboratory, Poona, however, does not confirm the analysis made

TABLE 1—CHARACTERISTICS OF TURPENTINE OIL FRACTIONS*

SAMPLE NO.	SP. GR.$^{30°}$	$n^{30°}$	$[\alpha]_D^{30°}$	YIELD OF α-PINENE %
I	0.8604	1.4625	−6.8°	77
II	0.8601	1.4623	−6.02°	71

* Examined by the Forest Research Institute, Dehra Dun.

TABLE 2—CHARACTERISTICS OF TURPENTINE OIL FRACTIONS*

FRACTION	TEMP. °C.	QUANTITY %	n
1	155-157	26.5	1.4680
2	158-160	40.4	1.4708
3	161-163	21.4	1.4741
Residue		10.4	. .
Loss		1.3	. .

* Examined by the National Chemical Laboratory, Poona.

TABLE 3—FRACTIONAL DISTILLATION OF TURPENTINE OIL*

FRACTION	TEMP. °C.	QUANTITY %	SP. GR.$^{15.5°}$	$n^{25°}$	$[\alpha]_D^{20°}$
1	100-130	2.0	. .	. .	. .
2	154-156	74.0	0.8703	1.4721	−5.7°
3	156-158	14.0	0.8697	1.4735	−8.8°
4	158-160	5.0	. .	1.4737	. .
5	160 and above	5.0	. .	1.4885	. .
Residue	yellowish, sticky				

* Examined by the author.

TABLE 4—COMPARATIVE RESULTS OF FRACTIONATION OF P. KHASYA OIL FROM BURMA AND ASSAM

FRACTION	TEMP. °C.	BURMA %	ASSAM %
1	150-155	76.0	3.5
2	155-160	20.0	67.0
3	160-165	. .	24.0
Residue		4.0	5.5

at Dehra Dun. The figures obtained by the National Chemical Laboratory are: sp. gr.$^{30°}$, 0.8668 ; $n^{30°}$, 1.4718 ; $[\alpha]_D$, −6.93° ; acid val., 0.44 ; and ester val., 7.25. The data obtained on fractional distillation of the oil is given in Table 2.

The oil (76 g.) on treatment with hydrochloric acid gave **35** g. of pinene hydrochloride.

The properties of the oil examined by the National Chemical Laboratory were found to be very much similar to those of *P. longifolia* and different from those of *P. khasya* examined by other workers. It was suggested by the National Chemical Laboratory that there might have been some mistake in the identification of the pine trees from which the resin was collected.

The author after examining several samples of the turpentine oil derived from the pine resin of the Khasi and Jaintia Hills, obtained the following results: sp. gr.$^{15.5°}$, 0.8640 to 0.8700 ; n^{25}, 1.4730 to 1.4760 and $[\alpha]_D^{20°}$, −6.7° to −7.0°. The results of the study of the products of fractional distillation of the oil are given in Table 3.

The above results show that the oil from *P. khasya* from the Khasi and Jaintia Hills is a moderately good turpentine oil, but it differs from the oil of *P. khasya* from Burma. The comparative results of fractionation[1] of these oils are given in Table 4.

The study of the oil is being continued in co-operation with the Botanical Forest Officer of Assam.

The turpentine oil produced at the Assam Government Rosin and Turpentine Unit is being highly commended by the medical profession: the quality of the oil being very good for all purposes for which it is generally used. Marketing difficulties are, however, being felt for both rosin and turpentine due to the absence of sizeable paint, varnish and soap industries in the State.

Turpentine oil could be used as a solvent for the extraction of quinine and other alkaloids from cinchona bark. Preliminary experiments by the author in this direction have shown encouraging results. The turpentine oil, at suitable temperatures, quickly takes up as much of the alkaloids from the bark as 'quinsol' (a paraffin oil supplied by the Assam Oil Company, Digboi) generally used for the purpose, with the added advantage that it dissolves very little of the colouring matter present in the bark. This may be of special interest for the extraction of Assam cinchona bark with low (3.0 per cent) alkaloid content.

Agar oil from agarwood (*Aquilaria agallocha*)

Agar oil (Aguru) is obtained by the distillation of the fragrant dark brown resinous portions of the wood of *Aquilaria agallocha* (*sasi*), which grows mainly in the Garo Hills, Goalpara, Darrang, Nowgong, Sibsagar and the Cacher Divisions of Assam. It is also found, to some extent, in the foothills of Bhutan, in Tipperah and the Martaban Hills in Burma. Virtually, Assam holds the monopoly over this valuable wood. Healthy trees, however, do not contain agarwood. The dark brown oil bearing resinous patches in the wood are developed as the result of the attack of a certain type of fungus. The general indication of their presence being the diseased appearance of the top and side branches. Agarwood is collected only when the tree showing diseased appearance is either partially or completely killed.

The Forest Research Institute, Dehra Dun, at the instance of the Forest Department, Assam, investigated the problem of the extraction of the oil from agarwood. Based on the results of these investigations, a small agar oil distillation scheme was put up to the State Government. In the meantime,

TABLE 5—YIELD OF AGAR OIL FROM DIFFERENT GRADES OF THE INFESTED WOOD

GRADE	YIELD %	SP. GR.
Yellow wood	0.09	—
Wood dust	0.15	0.9934
Brownish wood	0.23	1.0075
Doom wood	0.40	1.0116
Superior doom wood	0.84	—
Superior brown wood	1.11	1.0080
Superior black wood	1.91	1.0118
Absolutely black agarwood (heavier than water)	2.19	1.0118

efforts in distilling the oil have been made by certain private agarwood dealers and the departmental proposals have been dropped.

Agarwood, on an average, contains about 0.3-0.5 per cent of essential oil. The oil content depends on the quality of the agarwood and the degree of resin deposition. The black resinous portions of the wood which are comparatively heavier, yield up to 3.0 per cent while the brownish yellow light wood, generally used for extraction, yields about 0.4 per cent of the essential oil.

The highly resinous black agarwood (*Krishna-Agaru*) is not used for the extraction of oil. It is sold as such for incense burning and other purposes and is exported to Iran, Arabia and Far Eastern countries. The yields from the different grades of the infected wood, obtained by the Forest Research Institute are given in Table 5.

Agar oil is a very thick oil of golden yellow to yellowish brown colour having a characteristic strong odour. It has a very high fixative value in blending perfumes and, as such, is in great demand. It is used mostly for making agar *attar* using sandalwood oil as base.

Assam produces on an average 500 maunds of agarwood of different grades per year. The ultimate cost of production of the oil depends on the collection cost of the oil-yielding grades. Most of the trees having agarwood in them are now found only in the interior forests. Experiments are being carried out by the Assam Forest Department to explore the possibility of artificial infection of 'sasi' tree plantations through multiple inoculation with suitable culture of fungus. If artificial infection proves successful, the production of agar, which is more or less a matter of chance at present, may be placed on a sounder basis. The species is being planted in suitable localities in the State.

TABLE 6—YIELDS OF CAMPHOR FROM O. KILIMANDSCHARIUM GROWN IN DIFFERENT LOCALITIES IN ASSAM

No.	Locality	Total distillate %	Camphor %	Camphor oil* %
1.	Darrang (Batasipur)	6.23	2.49	3.74
2.	Sibsagar (Hollngapur)	5.45	2.20	3.25
3.	do.	5.96	2.72	3.24
4.	do.	5.30	1.93	3.37
5.	Naga Hills (Rangapahar)	5.78	2.62	3.16
6.	do.	6.39	3.54	2.85
7.	Khasi & Jaintia Hills (Umling)	3.91	1.98	1.93
8.	do. (Nongpoh)	3.91	2.05	1.86

* The camphor oil still retains about 50 per cent of camphor of which 30-33 per cent can be recovered by fractional distillation.

Camphor from *Ocimum kilimandscharium*

This species of camphor-yielding plant is being cultivated on an experimental basis in different localities of the State and the Forest Department has a plan to increase the acreage with the availability of more facilities. The yield of camphor, so far, has been quite encouraging as shown in Table 6.

Essential oil from *Santalum album* (*Sweta chandan*)

The plantation of this species has been successfully tried in the Khasi and Jaintia Hills, Assam, and the heartwood from 8 year old trees has been found to contain 4.65 per cent of the essential oil. However, the formation of heartwood as compared to Mysore plants, was very poor.

Other essential oil-yielding plants found scattered in the State are lemongrass and screw pine (*ketaki*). Lemon and orange peels are also available and can be used for the extraction of citrus oils. However, no systematic investigation has so far been carried out on the availability of these materials and the nature and yield of essential oils obtainable from them.

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Some Aromatic Plants of Jammu & Kashmir

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A preliminary investigation of the flora of Jammu & Kashmir has revealed the presence of nearly a hundred aromatic plants in the State. About a dozen of them including *Acorus calamus, Angelica glauca, Juniperus macropoda, J. communis, Xanthoxylum alatum, Carum carvi, Tanacetum vulgare,* **and** *Sassurea lappa* **yield a fairly good percentage of volatile oil and can be economically exploited. About two dozen plants gave a comparatively lower yield while the remaining plants reputed for their aromatic properties yielded only traces of oil.**

The territories of Jammu & Kashmir State represent a wide range of physical and climatic variations. The maximum temperature in the shade in Kandi ilaqa at the foot-hills of Jammu (900 ft.) goes up to 120°F. whereas in Kashmir (5,000 to 15,000 ft. or more) it does not rise above 100°F. The wide range of climatic conditions favours the development of a rich and varied flora. Numerous drugs used in the British and other pharmacopoeia either grow here wild or are cultivated. Apart from a large number of wild-growing medicinal plants used in the Ayurvedic and Unani systems of medicine, a wide range of essential oil bearing plants is found throughout the State. A preliminary survey has revealed that about a hundred or more plants growing in this State yield aromatic volatile oil.

As a result of preliminary study it has been observed that some of the wild-growing aromatic plants including *Acorus calamus, Sassurea lappa, Angelica galauca, Juniperus macropoda, J. communis, Xanthoxylum alatum, Carum carvi, Tanecetum vulgare* yield a fairly high percentage of essential oil. The distillation material is available in considerable quantities and can be commercially exploited.

Acorus calamus Linn. (Sweet flag ; Vern.—*Bach yai*)

A semi-aquatic perennial herb with indefinitely branched rhizomes. It is a native of Europe and North America but has been cultivated in India in damp marshy places at 3,000 to 6,000 ft. above sea level. In Kashmir it grows on the borders of lakes and marshy areas. The rhizomes are used in indigenous medicine as emetic, nauseant, antispasmodic and carminative ; and also in the treatment of asthma, diarrhoea and dysentery. The normal oil of sweet flag is used as a perfuming material and also as a flavouring agent for gin and some types of beer.

The fresh and unpeeled rhizomes on steam distillation yielded a brownish yellow oil with agreeable odour. The characteristics of this oil along with those of Indian and foreign oil are given in Table 1.

The oil is not produced in India at present but the drug is available in considerable quantities and there is sufficient demand in foreign markets.

Angelica glauca Edgew. (Vern.—*Chora*)

Grows in abundance in moist places at altitudes of 7,000 to 9,000 ft., particularly in the Kishanganga valley in Kashmir. Locally the root is utilized as spice. It is also used in indigenous medicine for the treatment of dysentery and constipation, and as stimulant and cardiac tonic. Angelica oil obtained from *Archangelica officinalis* is used in perfumery and for flavouring liquors.

The dry roots of the plant were steam distilled and a brownish yellow oil obtained in 1.3 per cent yield showed sp. gr.$^{15^\circ}$, 1.0275 and n^{20°, 1.529 ; values reported in literature being: yield, 0.35-1 per cent ; sp. gr.$^{15^\circ}$ 0.859-0.918 and n^{20°, 1.477-1.488.

Archangelica officinalis Hoffm. var. *himalaica*

A large perennial erect herb assuming the size of a small tree, commonly found in Kashmir near the water channels at altitudes from 8,000 to 13,000 ft. especially in hills near Gulmarg and Gurez.

The fruit and the roots of the plant are reputed for their aromatic properties. In Europe, its essential oil is used as a flavouring agent in confectionery and liquors.

The fruits and roots collected from hills near Gulmarg yielded after drying 0.8 and 3.8 per cent essential oil respectively. The root oil has sp. gr.$^{15^\circ}$, 0.8922 and n^{20°, 1.4478 ; the values for fruit oil being: sp. gr.$^{15^\circ}$, 0.887 and n^{20°, 1.464.

This oil can be distilled in considerable quantity provided a fair demand for it is created.

Carum carvi Linn. (Caraway ; Vern.—*Zira*)

Caraway consists of dried ripe fruits of *Carum carvi*, an erect biennial herb indigenous to and cultivated in North and Central Europe, chiefly in Holland,

TABLE 1—CHARACTERISTICS OF ACORUS CALAMUS OIL PRODUCED INDIGENOUSLY AND IN FOREIGN COUNTRIES

	LOCAL OIL	INDIAN OIL	FOREIGN OIL
Yield (from unpeeled rhizomes), %	3.1	1.5-3.5	1.5-3.5
Sp. gr.$^{15^\circ}$	1.08	1.069	0.959-0.97
n^{20°	1.5475		1.5028-1.5078

and in Central Asia. In India, it is cultivated as a summer crop in the hills of Baltistan, Kashmir, Kumaon and Garhwal, at altitudes varying from 9,000 to 12,000 ft. In Kashmir the plant commonly occurs in Astore, Drawa, Gurez, Skardu and it is also cultivated in the valley. Caraway is largely used in both the Indian and Western systems of medicine as stomachic and carminative. Caraway oil is also used for flavouring wines and scenting soaps.

For the distillation of the essential oil, caraway fruit growing in Skardu, Gurez and Baghbanpura in Kashmir valley were procured. The characteristics of the oil are given in Table 2.

The caraway growing in various parts of Kashmir gives fairly good yield of oil which is up to B.P. standard.

The fruit is mostly used as spice and is sold at a comparatively high price of Rs. 13 to 15 per seer. No attempt is being made to distil the oil. If, however, the plant is cultivated on a commercial scale and the cost of production reduced, the oil can be prepared economically. There is a fair demand for the oil in India and abroad. At present most of the Indian demand is met by imports.

Hyssopus officinalis Linn. (Vern.—*Zufah yabis*)

Hyssop oil is mainly distilled from *Hyssopus officinalis* which is indigenous to the Mediterranean countries and Central Asia. The plant grows in Baluchistan and in the Himalayas at altitudes ranging from 8,000 to 11,000 ft. It is not indigenous to the Kashmir valley but has been successfully raised in nurseries.

The leaves of the plant are used in indigenous medicine as a stimulant, stomachic, expectorant, diaphoretic and as an emmenagogue. Hyssop oil is also much esteemed for flavouring purposes and is said to be an ingredient of some of the French liquors. The fresh and dry herbs gave on steam distillation a brownish yellow oil with aromatic odour in a yield of 0.36 and 0.7 per cent respectively. The oil showed sp. gr.$^{15°}$, 0.9375 ; and $n^{20°}$, 1.4778 ; values reported for the foreign oil being: yield fresh herb, 0.07-0.29 ; dry herb, 0.3-0.9 per cent ; sp. gr.$^{15°}$, 0.927-0.945 and $n^{20°}$, 1.473-1.486.

The yield of the local oil and its physical constants compare favourably with those of the foreign oil. The plant has acclimatized well for large scale cultivation in Kashmir valley and can be economically exploited.

TABLE 2—CHARACTERISTICS OF CARAWAY OIL DISTILLED FROM MATERIAL GROWN AT BAGHBANPURA, GUREZ AND SKARDU (KASHMIR)

	LOCALITY			
	Baghbanpura	Gurez	Skardu	B. P. Standard
Altitude, *ft.*	5,500	7,900	7,700	. .
Yield, %	4.3	6.8	8.5	3.5-6
Sp. gr.$^{15°}$	0.9095	0.8902	0.8907	0.910-0.920
$n^{20°}$	1.491	1.486	1.485	1.485-1.492

Juniper Berries

Juniper berries and the oil extracted from them have been used as digestive and diuretic remedies since ancient times. Juniper oil is much used for imparting to beverage 'gin' its characteristic flavour. Three species of juniper, *Juniperus communis, J. macropoda* and *J. recurva* grow in Kashmir..

Juniperus communis Linn. (Vern.—*Vither*)

An evergreen shrub which grows at high altitudes—9,000-14,000 ft.—in the inner dry ranges of Kashmir. The ripe berries, procured from the Sindh valley, when dried and steam distilled yielded 0.77 per cent of a pale yellow oil having sp. gr.$^{15°}$, 0.9388 and $n^{20°}$, 1.488. The values mentioned in literature are: yield, 0.5-1.05 per cent ; sp. gr.$^{15°}$, 0.865-0.890 and $n^{20°}$, 1.475-1.488.

Juniperus macropoda Boiss. (Vern.—*Padam*)

A medium-sized tree which grows in the inner dry ranges of the Himalayas at altitudes varying from 8,500 to 14,000 ft. It is very common in the Gurez valley. The dry berries from Gurez yielded 4.4 per cent of a sweet smelling oil which has sp. gr.$^{15°}$, 0.8571 and $n^{20°}$, 1.473.

Saussurea lappa (C. B. Clarke ; Vern.—*Kuth*)

A herbaceous plant growing in Kashmir forests in the Kishanganga valley and the higher elevations of the Chenab valley. It is also found at higher elevations in Reasi, Ramban and Udhampur Divisions.

The roots collected in autumn before snow-fall, are mostly used as stimulant, and in the treatment of cough, asthma, fever, dyspepsia and skin diseases. Locally it is employed as a preservative for woollen goods. Large quantities of this drug are annually exported to China where it is burnt as incense in pagodas.

The roots, collected locally and dried, on steam distillation, gave 0.54 per cent of a pale brown oil having sp. gr.$^{20°}$, 1.051 and $n^{20°}$, 1.499.

Tanacetum vulgare Linn. (Tansy)

A perennial aromatic herb indigenous to and extensively cultivated in Europe. The flowers are yellow, the heads being in terminal corymbs.

The plant does not grow in India but it has been found growing as an escape near Yarikah in Kashmir. It may have been introduced as an ornamental plant. It is hardy and can be easily cultivated on an extensive scale.

In Europe the plant yields 0.1-0.4 per cent of volatile oil which is used as a flavouring agent and contains thujone, borneol, camphor and resins.

The whole flowering herb yielded 0.6 per cent of an essential oil with sp. gr.$^{15°}$, 0.939 ; $n^{20°}$, 1.4615 ; the values for American oil being: yield, 0.2-3 per cent ; sp. gr.$^{15°}$, 0.925-0.935 and $n^{20°}$, 1.457-1.459.

Xanthoxylum alatum Roxb. (Vern.—*Timbru*)

A shrub, at times attaining a height up to 20 ft. It grows along the foot-hills of the Himalayas from the Indus eastwards ascending to an altitude of 5,000 feet usually in hot dry places. In the State, the plant is common in Jhelum valley and in the foot-hills of Jammu.

Every part of the plant possesses a peculiar aromatic pungent smell. In indigenous medicine the fruits and bark are recommended in fevers, dyspepsia and cholera.

The fruit on steam distillation yielded 2.1 per cent of a pale yellow oil with a peculiar odour having sp. gr.$^{15°}$, 1.025 and $n^{20°}$, 1.475 ; the values for foreign oil being: yield, 3 per cent ; sp. gr.$^{15°}$, 0.8653 and $n^{20°}$, 1.4813.

TABLE 3—AROMATIC WILD PLANTS OF JAMMU & KASHMIR, YIELDS OF THEIR ESSENTIAL OILS AND COMPARATIVE DATA FOR PLANTS GROWING ABROAD

PLANT	PART DISTILLED	YIELD FROM KASHMIR PLANT %	YIELD FROM FOREIGN PLANT %
Arictum lappa	roots	0.75	. .
Abies pindrow	needles	0.61	. .
Cinnamomum tamala	leaves	1.2	. .
Chaerophyllum villosom	fruit	0.98	. .
Elsholtzia crestata	whole	0.93	2.0
F. jaeshkenna	roots	1.2	. .
Ferrula jaeshkenna	seeds	3.8	. .
Heracleum cashmiriana	roots	0.8	. .
Mentha sylvestrus	leaves & flowering tops	1.2	0.9
Moringa longifolia	whole	0.34	. .
Napata cilliaris	whole	0.54	. .
Ocimum sanctum	leaves & flowering tops	0.9	0.60
O. bascillicum	leaves & flowering tops	2.3	. .
O. canum	leaves & flowering tops	0.5	0.65
Prangus publaria	leaves & flowering tops	0.65	. .
Pinus longifolia	needles	0.22	. .
P. excelsa	needles	0.38	. .
Picea morinda	needles	0.22	. .
Senecio jackmotianus	roots	0.94	. .
Skimmia lauriola	leaves	0.45	. .
Thymus surphyllum	whole	0.72	0.15-0.6

WILD GROWING PLANTS

Besides these, a large number of aromatic plants growing wild in the State was investigated and a list of them together with the percentages of essential oils obtained is given in Table 3. They have not been described in detail because their oils have not found appreciable use in perfumery or medicine. These plants are available in sufficient quantity, and if a detailed investigation is carried out, it is likely that some of them may prove useful in the pharmaceutical and cosmetic industry or in the manufacture of insecticides and similar products.

In addition to the above, a number of plants which are reputed for their aromatic properties and grow wild in the State were studied but they yielded only traces of essential oil. These are: (1) *Achillea mellifolia*, (2) *Aegle marmelos*, (3) *Anethemus Noblis*, (4) *Artemisia dracuculus*, (5) *A. amygdalis*, (6) *A. grata*, (7) *A. perviflora*, (8) *Betula utilis*, (9) *Cauclais latifolia*, (10) *Capparis spinosa*, (11) *Elscholtzia densa*, (12) *Eryngium caeruleum*, (13) *Innula recemosa*, (14) *Iris kashmiriana*, (15) *Rhus succidane*, (16) *Macrotima benthame*, (17) *Mentha viridi*, (18) *Nepeta ruderalis*, (19) *Nepeta erecta*, (20) *Origanum vulgare*, (21) *Plectranthus rigosis*, (22) *Saliva moorcraftiana*, (23) *S. glutinosa*, (24) *S. dumetorum* and (25) *S. hians*.

A few species of rose and jasmine grow wild and are also cultivated in the State. *Jasmine sambricum* (motia) cultivated in the foot-hills of Jammu Province, is well known for its sweet fragrance in northern India. Flowers of rose and jasmine have been exploited only on a limited scale in the State for preparation of rose oil, rose water and jasmine oil.

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Potentialities of Essential Oil Industry in North Kanara District

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North Kanara is one of the most suitable areas for development of the essential oil industry. Cinnamon leaves, lemongrass, *khus* (vetiver) roots, *jummi* (*Zanthoxylum rhetsa* and *Z. budrunga*) fruits and *suringi* (*Ochrocarpus longifolius*) flowers are available in large quantities and may be used for the distillation of essential oils.

Essential oils are the odoriferous steam volatile constituents of plant materials. They are important products, so much so that they are looked upon as strategic materials[1]. There is a great scope for the development of essential oil industry in North Kanara. North Kanara is a narrow strip of land on the west coast of India with a rainfall of about 150 inches. It abounds in very rich forests which are yet untapped. As the soil and weather conditions in this area are similar to those in South Kanara, and Travancore-Cochin, the essential oil industries thriving in those regions can as well be developed in this district. The production of essential oils from the plant materials available in large quantities is discussed below.

Cinnamon oil—In 1944 India imported 17,000 lb. of cinnamon bark and leaf oils from Ceylon[2]. The quantity of cinnamon leaf oil imported in 1946 was 37,312 lb. In eugenol content, cinnamon leaf oil is at par with clove oil, largely used by the manufacturers of food products in America.

Cinnamon trees are found in large numbers in the forests of North Kanara. A survey in 1940 showed that nearly 1,00,000 lb. of leaves could be collected. A preliminary study conducted recently by the author has revealed that nearly 3,00,000 lb. of cinnamon leaves are available. With an average yield of about 0.7 per cent of oil, about 2,000 lb. of cinnamon oil may be produced in this area.

Lemongrass oil—Lemongrass oil ranks second among the essential oils exported from India. It is obtained from lemongrass grown mainly in the northern parts of Travancore State. India exported 1,22,469 gallons of oil during the year 1951-52; the export during 1952-53 was only 92,092 gallons. The oil is exported chiefly to England and America where it is extensively used in perfumery, and as a source of citral.

The soil and weather conditions in the downghat region of North Kanara are ideally suited for the cultivation of lemongrass. The average annual growth of grass in this region is about 20,000 lb. per acre, and the same yield can be expected of lemongrass also. The yield in Travancore State is 15,000 lb. per acre[3].

The author has been experimenting on the cultivation of lemongrass at Kumta from the seeds obtained from the Lemongrass Research Station, Odakkali in Travancore-Cochin State. The growth of the grass is quite satisfactory. Studies on the extraction and chemical composition of the oil are in progress.

Khus oil—*Khus* grows wild on the banks of the Varada river. It is found in a few places in Kumta and Honavar talukas. The grass in this district belongs to the non-flowering variety. If properly manured, it develops long thick roots yielding as much as 0.4 per cent of oil, whereas the roots from poor soils yield less than 0.1 per cent. The colour of the *khus* roots is mostly light yellow. It is reported that about 100 acres are under cultivation. The cultivation of *khus* and the extraction of oil from its roots will be a profitable enterprise.

Suringi (Ochrocarpus longifolius) *oil*—*Suringi* flowers are available in large quantities in North Kanara, and the author wishes to draw special attention to them. The flowers possess an extraordinarily pleasant aroma resembling that of *nagakesara* (*Mesua ferrea*). *Suringi* has not received the attention it deserves ; if properly developed, *suringi* oil will definitely prove to be an important and useful essential oil.

Jummi (Zanthoxylum rhetsa *and* Z. budrunga) *oil*—A large number of trees of this group are found throughout North Kanara district. The seeds yield about 0.8 per cent volatile oil containing *l*-sebanine. It is a yellowish oil with a peculiar aroma.

The oil and the seeds are known for their beneficial physiological activity[4] and may prove to be a useful ingredient in the preparation of food products.

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Development of Vetiver Oil Industry in South India

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It has been established that vetiver can be profitably grown as a rain-fed crop in the waste coastal lands of south west India. Laterite loam is the best soil and the roots take 15-18 months to mature. Heavy rains before harvest reduce the yield of oil which is also diminished by drying and storage of roots. The North Indian variety of seed setting vetiver has been advantageously introduced in the south.

South India, which was hardly producing any vetiver oil during pre-war years has built up a progressive vetiver oil industry capable of producing 500 to 600 lb. of oil annually. The demand for vetiver oil is likely to increase by 3 to 4 times allowing a great scope for further expansion. This industry has a special capacity to employ agricultural labour, as over 80 per cent of the cost of production of the oil is accounted for by labour charges.

The Indian vetiver (*Vetiveria zizanioides* Linn.) has the reputation of being one of the finest among oriental perfumes. In India it has been used in medicine and perfumery since the earliest period of her history. There are numerous references to this plant in the ancient Ayurvedic treatises by Charaka, Vagbhadananda and other writers. The copper plate inscriptions found near Etawah in Uttar Pradesh, reveal that the King of Kanauj took advantage of a flourishing trade in *khus* and levied an import duty on this commodity as early as 1103 A.D.

Occurrence—In India vetiver occurs under a variety of conditions ranging from sandy sea coasts and swamps to hill tops. It grows wild on the east and west coasts of India, in the waste lands of Orissa, in Bharatpur and other parts of Rajasthan, in the Punjab and Kumaon ascending to an altitude of about 2,000 feet. It is also found in Java, Malaya, Reunion and Seychelles islands, Louisiana (U.S.A.), Uganda and Gold Coast areas of Africa, Fiji, Philippines, Jamaica, Mexico, Honduras, Haiti and Guatemala.

INDIAN INDUSTRY

Although cultivation and distillation of vetiver oil have attained high proficiency as an industry outside India especially in places like Java, Reunion, Haiti and Seychelles, the Indian distillers appear to be satisfied by merely digging out whatever roots could be obtained from the wild growth. Sirsa and Kaithal in the Punjab, Bharatpur in Rajasthan and Musanagar, Utripura, Sitapur, Gonda and Kheri in Uttar Pradesh are reputed centres for wild *khus* roots. Its cultivation has been practically unknown in India till the last decade, except for small areas of coastal sandy soil of Ponnani in south Malabar and parts of south Travancore where some sort of shift cultivation has been in vogue for a long time. The roots grown are utilized for making hot weather *tatties* and fans for export to north Indian centres and also for use in the indigenous materia medica. As the oil obtained from the wild roots was insufficient to meet the increased demands of soap and cosmetic industries, India was obliged to import large quantities of vetiver oil from Java, Reunion, Seychelles, Haiti and other countries. With the occupation of major vetiver growing areas by the Japanese in 1941, the import of vetiver oil into India came to a sudden stop and the country was faced with the problem of improving its own resources for supplying the minimum requirements. As vetiver oil, in addition to being a highly valued perfume by itself, has an intrinsic value as a perfume fixer, the soap industry found it almost impossible to dispense with the use of this oil or to discover a satisfactory substitute for it. The prewar price of Rs. 30-35 soared to Rs. 160-180 per pound at the end of the war.

It was with this background that the Essential Oils Research Committee of the Council of Scientific & Industrial Research decided to undertake a survey of the Indian vetiver industry for exploring the possibilities of future development in various parts of the country. The committee appointed Shri C. T. Ittichan to undertake the survey, which ultimately resulted in the publication of the Monograph on Indian Vetiver.

The Industries Department of the Government of Madras decided to undertake commercial cultivation of this crop under the auspices of the Kerala Soap Institute, Kozhikode, utilizing the fairly large areas of vacant land attached to the various fish curing yards along the west coast. The author was appointed in July 1943 to undertake the cultivation which was started in August-September 1943. It was the first step in the regular and scientific study of this crop in India.

Cultivation—Though the cultivation was started rather late in the season and with little knowledge, even with some misconceptions regarding the requirements of the crop with respect to soil, manures and maturity, the area progressively increased from about 6 acres in 1943 to 30 acres in 1945. As it was spread over a number of fish curing yards distributed over the two districts of Malabar and South Kanara, the facilities for close observation and study were naturally limited. However, some valuable observations were recorded on several aspects of raising vetiver crop.

Generally the soil at all centres was of the white sandy type. Only at Tanur and West Hill near Kozhikode, there were a few small patches of a mixture of laterite and sandy soils. The roots grown in these patches were found to be thicker and hardier. On distillation they gave higher yields of oil than the thin hairy roots obtained from the sandy areas. The original idea that sandy soil is the best soil for vetiver appeared for the first time to be incorrect. By successive distillations it was established that vetiver roots produced in sandy soils, though quite soft and suitable for making fans and *tatties*, are very poor in oil content. Their yield of oil ranged from 0.2 to 0.3 per cent, while it was 0.75 to 1.00 per cent for the roots grown in laterite loam. The disparity in oil content is so great that it completely rules out sandy soils for the cultivation of roots intended for distillation. It has been found that for obtaining the maximum yield of oil, the period of optimum maturity for the rain-fed crop is 15 to 18 months.

The roots for distillation should not be harvested either during heavy rains or immediately after the rainy season, as such roots are found to be poorer in oil content compared with the roots of same maturity harvested in the dry season.

The application of concentrated manures like ammonium sulphate, groundnut cake or fish refuse tends to increase the yield of roots as well as their oil content.

The Kerala Soap Institute did not possess suitable land for carrying on the cultivation and therefore the scheme was terminated in 1946. A few private individuals headed by Shri D. V. Deo of Cochin, however, came forward to take up commercial cultivation and distillation of vetiver oil. They soon brought into existence an estate of about 30 acres at Nilambur in south Malabar. Cultivation which was started with about 10 acres in 1948 has progressively developed. At present the crop covers an area of about 50 acres, of which about 25 acres are at Nilambur and 20 to 25 acres at Elankoor near Manjeri, where a new block of about 350 acres has been purchased mainly for the expansion and development of vetiver cultivation. The annual production of oil in the two estates is estimated to be about 450 lb. valued at nearly Rs. 35,000 to Rs. 40,000 as against practically nothing in south India 10 years ago. The price of the oil which rose to Rs. 170 to Rs. 180 per lb. during the years of scarcity is gradually coming down.

In addition to the expansion of the cultivation and the distillation industry, the trials conducted by the cultivators have also yielded valuable information. It has been shown that roots distilled immediately after harvest yield about 25 per cent more oil than the roots distilled after drying and storing, though there is practically no difference in the quality of the oil. This corrects the old notion that storage of roots improves the quality of oil.

North Indian vetiver—A hundred slips of Bharatpur vetiver were specially obtained from the Divisional Forest Officer, Bharatpur, in 1950 and planted at Nilambur. Out of them only about 60 sprouted and flowered, and were found to set seed as discovered by the appearance of a large number of seedlings under the plants during the next rainy season. These seedlings as well as the available slips obtained from the 60 plants which survived have been very carefully used for multiplication of plants and extension of the area under cultivation. The South Indian vetiver, though it flowers profusely, does not set seed. Its propagation is limited to the more costly vegetative method, whereas in the case of the north Indian variety the seeds shed by the plants germinate and completely replace the harvested crop. The digging out of the roots serves as a sort of preparatory cultivation for the self sown seeds. In the south Indian variety, a crop once harvested is lost for ever unless the slips are carefully preserved and planted again.

The first distillation of the north Indian vetiver, which now occupies an area of about 10 acres at Elankoor, was carried out in 1952-53. The oil obtained had a special aroma quite distinct from the oil of the south Indian variety, though both the oils are considered equally valuable by the trade. The oil from the south Indian vetiver is found to be dextrorotatory while that of the Bharatpur variety is laevorotatory.

There is a definite preference by the soap industry for the oil obtained from the north Indian variety of vetiver. This variety, because of its self propagation through seeds, is particularly valuable when planting is done on hill slopes and other places which are difficult to plough. There is, therefore, a fair chance of this variety completely replacing the south Indian type in course of time so far as cultivation for oil distillation is concerned.

As the minimum requirement of vetiver oil by the Indian soap and cosmetics industries is estimated to be about 5 tons per annum and the present Indian production amounts to only about 1½ tons, there is a wide scope for the expansion of this industry especially in the South. The offtake of the oil is likely to increase to three or four times the present quantity when the oil becomes available at cheaper rates, since it is only its prohibitive price which is forcing the industry to go in for cheaper substitutes. As the cost of labour comes down with the return of normal conditions, it might be possible to produce and supply the oil at considerably lower rates. When the planting of slips is replaced by suitable spacing of self sown seedlings of the north Indian variety, a further substantial reduction in the cost of production is bound to result, especially when it is remembered that the cost of preparing the land, laying ridges and planting, and the cost of slips required, account for nearly 25 per cent of the cost of production of the oil[2].

The analysis of a representative sample of oil from south Indian vetiver

TABLE 1—ANALYSES OF SOUTH INDIAN AND BHARATPUR VETIVER OILS

SOURCE	YIELD OF OIL %	COLOUR & AROMA	SP. GR.	n	$[\alpha]D^{30°}$	ACID VAL.	ESTER VAL.	SOL. AT 25°C. 80% ALC. VOL.
Nilampur (S. Indian)	1.20	Brownish yellow with fine *khus* note	0.9870	1.5242	+16.5°	13.4	40.93	1.2
Bharatpur[3] Baraha	0.88	Deep brown viscous fine *khus* note	0.9942	1.5164	−64.2°	12.4	19.80	1.1
Bharatpur[3] Borai Centre	0.90	Golden yellow viscous, spicy *khus* note	0.9960	1.5172	−83.0	12.0	23.50	1.1
Bharatpur[3] Borai Centre	1.17	Reddish brown light, true *khus* note	0.9964	1.5170	−51.0°	14.5	26.40	1.1

produced at Nilambur with corresponding figures for the oil from Bharatpur variety is given in Table 1.

CONCLUSION

It has been proved that vetiver can successfully and profitably be cultivated as a rain-fed crop in the vast waste lands of the west coast of India.

The north Indian or Bharatpur variety of vetiver has been successfully introduced into the South. This variety on account of its capacity to set seeds and produce self-sown crops has a definite advantage over the local south Indian variety.

South India which was hardly producing any vetiver oil during the prewar days has built up a progressive industry on the West Coast which is capable of producing 500 to 600 lb. of oil costing Rs. 40,000 to Rs. 45,000.

The present Indian production of vetiver oil is sufficient to meet only about 30 per cent of the minimum requirements of the Indian soap industry. When prices are reduced with the fall in the cost of production, the demand for the oil is expected to increase three or four times, and there appears to be a great scope for the development of this industry in the south.

As over 80 per cent of the cost of production of oil is accounted for by labour charges, this industry has a special capacity to employ a large number of agricultural labour.

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Lemongrass Oil Industry in South India

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A general survey of the defects and deficiencies at various stages of lemongrass industry has been made. While suggesting methods to overcome them, emphasis has been laid on the co-operative organization of production and marketing, quality control at various stages from cultivation to the export of oil and the utilization of lemongrass waste.

South India used to lead the world market of lemongrass oil both in quantity and quality. But lately, this dollar-earning cottage industry has been deteriorating on account of unscientific cultivation of quality lemongrass, crude distillation process, unorganized marketing, lack of quality control at various stages, adulteration in trade and the absence of proper Governmental control and encouragement for improvement.

The plantations of lemongrass are on small land holdings of individuals and are not well looked after. When the price of lemongrass oil appears to be desirably lucrative the cultivators pay attention to their plantations. When the price is low the plantations practically remain abandoned. Weeds grow among the good grass, which suffers in health and quality. Cultivators, interested in immediate profit only, plant a mixture of white and red varieties of lemongrass in order to get more oil with the minimum required percentage of citral. These plantations not only yield a low quality oil but also in course of time produce a different variety of grass by cross pollination. Seeds taken from such plantations are being used for fresh plantations and thus the original good variety of grass is being gradually eliminated. Though unknowingly, a great harm is thus being done by the cultivators to the industry.

This practice should be stopped by a strict control of the seeds. It is really a great problem for the sensible and sincere industrial lemongrass cultivators to collect good seeds or seedlings for a fresh plantation. As the success of the industry depends on the correct quality of the grass planted, the Essential Oils Research Committee, CSIR, with the co-operation of the Government Lemongrass Stations should distribute good quality seeds at reasonable rates to the cultivators. Wide publicity should be given in the lemongrass centres to the harmful effects of using indifferent quality of seeds and seedlings.

Local committees formed at various lemongrass centres on co-operative basis should do their best to eradicate the low quality grass and weeds from all fields and their vicinity within their jurisdiction. Small financial help to deserving cultivators should be rendered by these committees with a view to improving the quality of every drop of oil produced in their area. As lemongrass fields are scattered and the cultivators are poor, active co-operative committees with Government backing are required to ensure that the lemongrass fields are properly weeded and kept in good condition perpetually. The 'white grass' can be called 'demongrass' and should be fought as such.

Distillation—The type of distillation unit now in general use in South India is very antique and crude. Copper stills and copper condensers are being used at present. A change of the condensers from copper to aluminium will show a great improvement in the colour and clearness of the oil. A leak-proof still, and a perfect separator with a back-flow arrangement for the water to flow from the separator to the water chamber will increase the yield of oil. Control of distillation time will increase the citral content and reduce the fuel consumption with the consequent reduction of the cost of production. Insulation of the still and a portion of the 'goose-neck' connecting it with the condenser will result in quicker and more efficient distillation. It may be pointed out, on the basis of practical experience, that oil distilled in a scientifically improved distillation unit was sold at 20 per cent higher price than the prevailing market rate in foreign countries even at a time when the demand for lemongrass oil was very poor. Leading lemongrass oil exporters including foreign firms like Messrs *Volkart Brothers* and *Aspinwall & Co.* at Cochin felt that the oil so distilled was chemically processed and rectified. By simple modification of the present distillation unit, the quality of oil could easily be improved and brought up to a considerably high standard.

Marketing—Cultivation and distillation are comparatively easier for small cultivators and distillers of lemongrass than selling the oil. The first step of adulteration takes place knowingly or unknowingly in the process of cultivation and distillation. The country distiller who wants to earn his day's bread by disposing off the day's product sells his oil to a local small purchaser who takes more in measure and pays less in price. Before this product reaches the final exporter, it passes through many intermediary traders all of whom share a small profit and also mix all sorts of adulterants to raise or lower the final citral content of their stock. It is mainly in the process of marketing that the good oil suffers in quality. The poor cultivator or distiller has to depend on financiers to carry on with his trade and is consequently compelled to sell his product to the financier at a price fixed by the latter.

It is very essential to form local co-operative marketing societies to control the quantity as well as the quality of the oil produced in each lemongrass centre. These co-operative marketing societies, amalgamated with the local lemongrass committees can be controlled by a central co-operative marketing society which could ensure that each distiller and/or cultivator gets at least his cost of production with a reasonable profit for his product even when the market price falls low. If there exists an assurance of this nature, the cultivators and distillers will take more interest in their fields and produce and keep up the quality of the oil and status of the industry. The central marketing society can control the export and ensure that each drop of oil going out of the country is of the best quality.

The marketing society with co-operation of the Government Research Stations can also contribute much to the improvement of the industry. In every industry attempts are being made to utilize the waste materials and prepare by-products. No practical use worth mentioning is being made of the refuse lemongrass left after distillation. It is a good manure. While the agriculturists do not get sufficient quantities of good manure for their fields, large amounts of it are going to waste. It is not even returned to the lemongrass fields. Large quantities of straw are being used as packing material. Whether it is possible—to (*i*) use the dried refuse grass in its place and save the straw for the cattle, (*ii*) mix the waste grass with molasses and chemical binder and preserve it as cattle fodder, (*iii*) briquette the waste and use it as fuel for distillation and (*iv*) use it as a raw material in paper and cardboard industry—are problems which have to be solved by research institutions. The central marketing society may be instrumental in solving these problems and enable cultivators and distillers to get a little extra income.

Quality Control—Control of the quality of oil at the port by the Quality Control Station by only passing good oil is not very conducive to the welfare of the industry. Small dealers in the oil at villages will take advantage of the rejection of oil of poor quality at the port to condemn the oil produced by the small distillers or pay low price for the oil. Besides, it is not possible for the small distiller to supply free samples of oil for test and pay the bill for testing and accept the decision of the purchaser as to the quality and price of the oil. Hence, quality control should start at the stage of supply of seeds and seedlings and through every step of production and marketing up to the stage of export.

Place in India's Five-Year Plan—Nearly 80 per cent of lemongrass oil exported from the country is produced in South India. While schemes of the Five-Year Plan are worked out in this region due regard should be given to this industry. The Chalakudi Community Project area in Travancore-Cochin State includes vast areas of lemongrass fields and a good scheme for the development of this industry could be worked out under this Project. A demonstration farm, mobile demonstration distillation units, nursery farms, educative propaganda programmes for the village distillers and cultivators, co-operative cultivating societies and co-operative marketing societies could be organized under the community project and worked out successfully to the benefit of the industry. A small committee could collect statistics of lemongrass fields and owners in various parts of Travancore-Cochin State and Malabar. It will be helpful in preparing suitable development schemes. In Wynaad and other parts of Malabar, cultivation of lemongrass has been expanded by enterprizing individuals, but due to lack of proper organization of the industry in general the progress has been very slow.

Development of Lemongrass Oil Industry in South India

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Lemongrass oil industry is an important dollar-earning agro-cottage industry of North and Central Travancore-Cochin and South Malabar providing employment to thousands and producing about 80 per cent of the world's total output of this oil. In order to further develop the industry, it is suggested that, on the one hand, more advice and better planting material should be made available to the ryots, and on the other, a better method of distillation may be popularized through distillation demonstration centres. A central refinery may be established for the production of standard refined oil for export. Co-operative production and marketing should be encouraged and better uses of the oil and the grass waste may be developed. Complementary cultivation of other essential oil bearing plants may be explored in order to utilize the idle distillation equipment during the summer.

The quantity and value of lemongrass oil exported from 1934 to 1953[1] are given in Table 1. This industry involves thousands of ryots, labourers and small businessmen and requires systematic development for a more profitable exploitation of our lemongrass resources.

PRESENT POSITION

The production of lemongrass oil is more or less monopolized by the Travancore-Cochin State. Formerly, only wild grass was used for the distillation of oil but in course of time, the ryots, with a view to improve the quality of the oil and to decrease the collection charges, began to cultivate the grass. The producers in the area use primitive methods of distillation and the small quantities of oil obtained are sold through field brokers to village merchants. Through the usual channels of trade the oil finally reaches the shipping port at Cochin. The lemongrass oil produced in this area is renowned for its high citral content and is commercially known as "Cochin oil" or "East Indian oil". With the increase in demand in foreign markets, the area under cultivated lemongrass has increased enormously. At present, more than 40,000 acres in South India are under this grass and more than 700 tons of oil (about 80 per cent of the world's total production) are being produced.

Plantations

The lemongrass oil industry is concentrated in the midland regions of North and Central Travancore, eastern taluks of Cochin, and in some parts of South

TABLE 1—EXPORTS OF LEMONGRASS OIL FROM INDIA

Period	Quantity gal.	Value Rs.
1934-35—1938-39 (av.)	90,213	9,18,226
1939-40—1943-44 (av.)	84,078	16,66,843
1944-45	121,629	32,12,243
1945-46	150,790	70,13,862
1946-47	132,290	107,95,131
1947-48	84,053	36,56,595
1948-49	95,824	24,07,677
1949-50	88,223	46,62,338
1950-51	127,706	132,64,366
1951-52	122,469	149,00,224
1952-53	92,092	39,17,100

Malabar. Extensive lands, where no other crops can profitably be grown, are available for the cultivation of this grass. Most of the plantings are located on low-priced hilly slopes. They vary in size and are scattered. A well-cared plantation lasts for more than seven years. The so called "red lemongrass" (*Cymbopogon flexuosus*), distinguishable by the colour of its stem, is extensively cultivated. It yields the genuine lemongrass oil containing 75 to 85 per cent of citral.

Field distillation

The production of lemongrass oil is characterized by the dispersion of small distilling units over widely scattered plantings mainly due to the difficulties in transportation. A centrally located distillery does not exist. The scene of operation is a small thatched shed with a crude still which is a small portable contraption, low-priced and easy to operate.

The distilling unit generally consists of a retort about 4 ft. high and 7 ft. in circumference and has a capacity of about 125 gal. The retort and the condensing spiral are made of thin plates of copper. At one side, about 2 ft. above the bottom of the retort, is provided a man-hole through which the plant material can be charged and discharged. The grass mixed with water is placed in the retort and it is heated over naked fire. During distillation the man-hole is closed with a cover held fast by means of cowdung which hardens on drying. The operation follows the principle of water distillation. The volatile oil mixed with steam goes through a goose neck into the spiral condenser inserted in a large wooden barrel containing cold water. The receiver is a wide beaker fitted with a syphon tube at the bottom facilitating the discharge of the condensed water, leaving the oil behind. Distillation of one charge takes nearly 3 hours. The yield of oil varies with the quantity of charge, nature of the material and climatic conditions. The oil is skimmed off from the collecting vessel by an experienced labourer and is stored in bottles.

The still tops are not removable and the task of charging and discharging through the side is laborious.

Defects of field distillation

The defects of the present method of field distillation, such as the tedious task of charging and discharging, long duration of single distillation, wastage of oil and fuel, and decomposition of the oil probably due to over-heating and charring are many. The small distillers do not observe the fundamental principle of water-distillation, that is, small plant charge and quick distillation. The ryots charge their stills as high as possible in order to utilize them fully and the speed of distillation is limited by a small condenser. There is no arrangement for continuous circulation of cold water in the condensing system and the distillate usually runs warm. A part of the oil remains emulsified or suspended in the warm water. The separation of oil and its storage is also crudely done.

DEVELOPMENTAL ASPECTS

The development of this agro-cottage industry involves two important aspects: A better cultivation of the grass and an improved method for the extraction of oil. Systematic agricultural research has to be carried out in order to select the proper variety of grass, yielding the maxmium quantity of quality oil. Advice regarding the optimum conditions of soil and climate necessary for the good growth of the grass should be made available to the ryots and arrangements should be made to distribute better planting material. This work is being undertaken by the Lemongrass Research Station started by the Government of Travancore-Cochin.

Distillation demonstration centres

The modern methods of distillation involve specialized skill, and a gradual step to modernize the distilling equipment should be attempted. As a preliminary measure 'distillation demonstration centres' equipped with improved modern stills should be established in typical areas of lemongrass industry. By intensive propaganda, the fundamentals of efficient distillation should be brought home to the ryots and the possibilities of installing steam distillation plants on co-operative basis should be investigated.

Refining plant

Pre-treatment of the oil before storage is not practised by the distillers and stockists. This omission is detrimental to the quality of exported oil. The oil distilled by the ryots is highly discoloured and contains much water, resinous matter and dirt. The impurities must be removed before export. It is essential that arrangements should be made to set up a central refinery for the purification of lemongrass oil.

Co-operative production and marketing of oil

The lemongrass oil industry of South India has been steadily declining. This is mainly due to the impure quality of the product and wide fluctuation in its price.

TABLE 2—LEMONGRASS OIL PRICE MOVEMENTS*

MONTHS	YEARS				
	1951	1952	1953	1954	1955
	Rs.As.	Rs.As.	Rs.As.	Rs.As.	Rs.As.
January	310-10	127-8	65-4	87-14	177-13
February	319-12	126-7	81-11	98-5	149-7
March	324-10	90-13	75-2	92-5	133-11
April	293-3	85-13	66-8	87-14	. .
May	265-7	82-5	65-4	84-1	. .
June	209-13	84-1	60-11	85-12	. .
July	198-2	79-1	57-6	94-15	. .
August	235-0	70-1	60-9	111-11	. .
September	210-13	65-10	62-5	152-8	. .
October	226-5	64-1	61-0	190-8	. .
November	197-11	60-0	66-8	182-8	. .
December	148-8	62-13	77-14	182-8	. .

* Average price for dozen bottles of 22 oz. each at Cochin.

TABLE 3—INDIAN STANDARDS* FOR LEMONGRASS OIL

Colour and appearance	Reddish yellow to brown ; mobile liquid
Odour	Lemon-like
Sp. gr. at 15°/15°	0.900-0.910
Sp. gr. at 25°/25°	0.892-0.902
Sp gr. at 30°/30°	0.888-0.989
$[\alpha]_D$	$-3°$ to $+1°$
n at 25°	1.4808-1.4868
n at 30°	1.4786-1.4846
Citral, by vol., %	75
Solubility, in alcohol, %	70

* IS: 327—1952.

The fluctuation in the price[2] of oil as given in Table 2 is due to the fluctuation in foreign demand and the monopolization of trade by some prominent merchants and stockists. As a remedy, an organization for the marketing of oil on a co-operative basis should be set up and a number of primary industrial co-operative societies at all important centres of the industry should be opened. The producers will sell the oil to the primary societies who will forward their consignments to the central society. The central society, which will be in touch with foreign markets, will export the oil. The rural industrial co-operative societies should be provided with adequate funds by the Government for advancing loans to cultivators for agricultural expenses and for installing new stills and mending old ones. The co-operative marketing system can only be introduced gradually. In the meantime, neighbouring grass cultivators should be encouraged to form themselves into groups and set up improved distilling units.

Quality control

Another serious drawback hampering the progress of this industry is the practice of adulteration of oil. This has lowered the reputation of our lemongrass oil in the foreign markets. Due to its doubtful quality, the demand for our oil has declined considerably. To prevent the export of adulterated oil, the Government of India have opened a Quality Control Laboratory at Cochin, where every consignment of oil is tested according to Indian standards and duly certified before export. Passing a quality control order and standardizing the packing of oil can attract foreign buyers. The present shipping contract for the oil should be modified so as to discourage adulteration. The Indian standards for lemongrass oil are given in Table 3.

Processing of lemongrass oil

Lemongrass oil is an important basic material for the manufacture of many aromatic substances used in soap, perfume, and cosmetic industries. The oil is exported in the raw state and even at peak prices the exchange earnings are very poor. Arrangements should, therefore, be made to manufacture aromatic products like ionone from the oil and research work should be undertaken for finding out alternative and better uses for the oil.

Lemongrass waste

At present the lemongrass waste is used as manure and fuel. Ways and means to utilize it in a more profitable manner have to be urgently investigated.

Cultivation of other essential oil bearing plants

The lemongrass distillers have to keep their distilling units idle for a part of the year, as the grass gets dried up in summer. Possibilities of the cultivation of other essential oil bearing plants like vetiver, patchouli and others, which can be distilled profitably, should be explored.

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ESSENTIAL OILS IN PHARMACEUTICAL INDUSTRY

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Essential oils, besides their use in perfumery, find application in the pharmaceutical industry because of their antiseptic, carminative, stimulative, expectorant, diuretic, counter irritant, rubefacient and flavouring properties.

The most important use of essential oils is in the manufacture of various types of syrups in which they are added mainly as flavouring agents and in the manufacture of spirits, e.g., spirity cinnamon, etc. and aquas, which are saturated solutions of the oils in water. When taken internally, they exert a mild irritating action on the mucous membranes of the mouth and digestive track, induce a feeling of warmth and increase salivation. Hence, they are used to stimulate appetite and aid digestion. They are excreted through the lungs, skin and kidneys and during excretion by the bronchioles they act as mild expectorants and are therefore included in cough mixtures, in inhalants, nasal and throat sprays, etc. In India, they find extensive use in gastric discomfort, flatulent colic and in mitigating the griping action of purgatives. Due to their antiseptic and rubefacient action they play an important part in the manufacture of lotions and ointments.

The use of essential oils in dentistry is based on their antiseptic, obtunding and stimulating properties. The oils commonly used are those of clove, peppermint, eucalyptus, cassia, etc. which produce severe irritation followed by anaesthesia.

Essential oils of pine, dill, anise, cinnamon, eucalyptus, lemon, peppermint, cajuput, sandalwood and turpentine are among the most important ones used in the English speaking countries, while in the Indian Pharmacopoeial List, 1946, essential oils of *ajowan*, orange flower, cassia, chenopodium, cinnamon leaf, cumin, fennel, gaultheria, lemongrass, juniper, *pudina* and turpentine are official.

With the exception of the oils of sandalwood, lemongrass, lemon, *ajowan* and turpentine, other oils, official in the Indian Pharmacopoeial List, are not prepared in appreciable quantities in the country. Consequently they are not available for dispensing and foreign substitutes have to be used. A proper development of indigenous essential oil industry will help the pharmaceutical industry in the preparation of galenicals, pharmaceuticals and ready-made medicines.

QUALITY CONTROL OF EAST INDIAN LEMONGRASS OIL

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Lemongrass oil forms an important perfume ingredient of a large number of household products of daily use like mouth washes, deodorants and liniment. Recent experiments carried out at Bangalore suggest that antiseptics of a fairly high Rideal Walker co-efficient could be prepared with lemongrass oil as the chief germicidal constituent. Standardization of its quality, therefore, is a matter of great importance for the manufacture of products involving its use.

The East Indian lemongrass oil is produced mostly on a cottage industry scale. Although it conforms to the British Pharmaceutical standard, particularly in its citral content, it exhibits a variety of colour even in samples taken from the same consignment. Colour is an important factor in a finished perfume or antiseptic. The oil obtained on cottage industry basis is very cloudy in appearance. It is necessary, therefore, to bestow proper attention to production of bright, clear, coloured oil conforming to definite standards in regard to colour, clarity, analytical constants and odour.

PRODUCTION OF AROMATIC CROPS AS FOREST UNDERGROWTH AND UTILIZATION OF FOREST WASTE LANDS

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Cultivation of essential oil bearing plants in idle forest lands in India is one of the effective steps in developing the essential oil industry in India and thus earning foreign exchange. Herbert Howard has estimated that well over 3 lakh square miles are available for this purpose.

A scheme costing 2-4 crores of rupees is suggested for developing 10 lakh acres of land for the culture of aromatic plants. It is estimated that this will result in an employment of 6 lakh persons and lead to an earning of a sum of Rs. 2.5 crores by way of foreign exchange.

USE OF GARLIC IN ANCIENT INDIA

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In sanskrit the most well-known names of garlic are *lashun*, *rashoon* and *rason*. It is difficult to fix up its native country. It has not been mentioned in the Vedas and ancient Hindu mythology. However, Sanskrit collections of medicinal literature conclusively show that garlic has been in extensive use in India for the last three thousand years both in culinary and galenical preparations. Due to its essential oil content, garlic has been a useful medicament in Ayurveda and has long been in use for the treatment of serious ailments of the respiratory, nervous and reproductive systems. Inhalation of its volatile oil has been recommended in cases of pulmonary tuberculosis, rheumatism, sterility and impotency. Because of its highly curative properties, it has been described as derived form ambrosia or *amrita*.

The later prejudice against the use of garlic in India appears to have originated from its popularity with foreign invaders. It was so intense that not only socio-religious writers like Manu deprecated its use, but also authors of medicine like Kashyapa, disparaged it.

The Essential Oil Resources of Mysore. M. N. Ramaswamy (Bangalore)

Indian For., **82** (1956), 127-129. Sandalwood oil, cinnamon (*Cinnamomum iners* Reinw.) leaf oil, and Indian lavender (*Bursera delpechiana* Poiss.) oil are produced in substantial quantities in Mysore and have established their reputation in world markets. The development has been due to a favourable climate, the impact of pioneering research on essential oils at the Indian Institute of Science, Bangalore, royal patronage of the indigenous perfumers and the work done at the Mysore Government Soap Factory. There is considerable scope for development of the oils from patchouli (*Pogostemon* spp.), vetiver (*Vetiveria zizanioides* Linn. Nash), peppermint (*Mentha piperita* Linn.), davana (*Artemesia pallens* Wall ex. Bess.) and camphor distillates from *Ocimum kilimandscharicum* Guerke.

The organization of the developmental work and the difficulties to be faced are discussed. The instances of essential oils from blossoms of *Shorea talura* Roxb. and fruits of *Zanthoxylum tetraspermum* Wight & Arn. are cited in support of the view that production of quality essential oil need not necessarily be in very large quantities to be commercially feasible. (*Abstract*)

Possibilities of Growing Camphor-yielding Ocimum in Madras State. C. Rajasekhara Mudaliar (Coimbatore)

Soap Perfum. Cosm. Tr. Rev., **29** (1956), 441. Cultivation trials of camphor-yielding ocimum (*Ocimum kilimandscharicum* Guerke.) have been carried out in Yercaud, Cinchona, Wynaad, Pattambi and Singampatti Hills (Madras State) with seeds obtained from the Forest Research Institute, Dehra Dun. The altitudes of the centres chosen for trial vary from 2,000-4,500 ft. The plants thrived well in Cinchona and Yercaud, while at Pattambi, their growth was not satisfactory. At Yercaud, a small quantity of camphor was obtained by distillation and found to be of good quality. It is interesting to note that the plant grew very vigorously in all waste places where no other economic plant will thrive.

It is suggested that the exploitation of this plant be taken up on a large scale in Cinchona and Yercaud. The Quinine Factory at Cinchona can take up distillation work and a distilling plant may be erected at Yercaud. (*Abstract*)

Cultivation of Essential Oil Bearing Plants and Some Connected Problems

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Cultural operations with reference to important essential oil bearing plants are discussed.

Since remote past, the essential oil industry has held a place of importance in Indian trade. In recent years the quantity of essential oils manufactured, especially from plants, has increased considerably. Standard specifications have been laid down for most of the oils, and large scale buyers are interested only in such oils as conform to the specifications.

Wildly growing plants have been the principal source of essential oils in India. These plants do not constitute an economic raw material for the production of essential oils of standard quality. To meet this requirement systematic cultivation of these plants has to be undertaken. India possesses areas of widely different climatic and soil conditions suitable for the cultivation of these essential oil bearing plants.

The essential oils occur in the cells of the plant body. In vetiver the fragrant substances are concentrated in roots; in *nagar-motha* (*Cyperus scariosus*), in rhizomes; in palmarosa and lemongrass, in leaves; in cinnamon, in bark; in rose and jasmine, in flowers and in coriander, dill and fennel, in fruits.

In systematic cultivation, we have to create conditions favourable to the healthy growth of the plant with particular reference to the parts bearing the concentration of essential oil. This involves a number of operations discussed below.

Preparation of the field—While preparing a field for the plantation of essential oil bearing plants, the type of ploughing will depend upon the type of plant to be grown. Root crops will require deep ploughing, while for foliage crops, shallow ploughing will be suitable.

In the case of palmarosa and lemongrass, it is necessary to plough the soil thoroughly, but only up to a depth of 4 to 6 inches. In the case of mints and ocimum careful weed free ploughing is advantageous. It should be followed by harrowing and disking. Initial weed free ploughing will ease successive weedings, and destroy the insect pests by exposing their hiding places; it will also remove the diseased vegetative matter from the soil.

In the case of vetiver, deep ploughing facilitates the emergence of a large number of roots, which penetrate deeper into the soil, resulting in increased yield. Deep ploughing makes it possible to reduce the distance between plants, as the roots instead of spreading sink deeper.

Sowing—(*i*) Spacing: The space allowed between row and row and plant and plant is determined by the nature of the plant and the type of soil. In the case of palmarosa and lemongrass, a spacing of 3 to 4 feet is sufficient for profuse growth. For mints, a spacing of 2 to 3 feet is the best; for vetiver, a spacing of 5 to 6 feet is necessary, as otherwise intercultural operations such as hoeing and weeding become difficult. This space may be utilized for growing inter-crops, when the plantation is young or harvested. (*ii*) Selection of rhizomes, root-stocks and seedlings: For raising a healthy plantation it is very essential to use healthy seeds. Rooted and diseased seeds should be discarded. For a standardized plantation only pure variety seeds should be selected ; this will minimise hybridization. It has been observed that hybridization of palmarosa varieties—*motia* and *sofia*—lowers the geraniol content in the essential oil. In the case of mints, hybridization may be advantageous as the hybrid variety is resistant to rust.

Transplantation—To obtain good results healthy selected seedlings are transplanted. It has been observed in vetiver that transplanted plants bear profuse vegetative growth and attain a height of 6 to 7½ feet as against 2 to 3 feet in wild growth. A good seedling gives a larger number of sprouts. The application of phosphatic manure after transplantation is helpful for the development of roots. In foreign countries this treatment has been found useful in the case of spearmint as well.

Irrigation—Vetiver is a very hardy plant, which withstands dry conditions in summer, but due to scarcity of water vegetative growth gets stunted. Summer irrigation induces early flowering; it also helps in raising suitable inter-crops of short duration. In the case of mints irrigation is more important for the development of succulent vegetative growth. Abundant moisture, with or without fertilizers, is required for a good yield of essential oil. Irrigation in winter protects the plantation from frost.

Drainage—Drainage allows the excess of water to flow away from the plant root system, and keeps the moisture content of the soil constant. It helps in maintaining the *p*H of the soil and improves its physical condition through aeration. It checks the process of denitrification and promotes the growth of beneficial micro-organisms, facilitating better root development and is beneficial to the essential oil content in root crops. In the case of mints over drainage should be avoided, as it reduces the optimum moisture content of the soil required by the plants for their proper development.

Manuring—Organic matter in sufficient quantity and nitrogen are very essential for the healthy growth of the plant. They should be supplemented by a manure which would particularly develop the essential oil bearing part of the plant. The application of phosphatic manures to vetiver in addition to nitrogenous fertilizers will help the development of roots. It has been observed in south India that the application of brine manure, groundnut cake and ammonium sulphate increased the oil content of vetiver. As the essential oil is contained in the parenchymatous tissues between the epidermis and endodermis, the development of root in size becomes necessary for the enhancement of oil content. In the case of plants like mint, fertilizers with high percentage of potash are recommended. It is reported that the application of sodium nitrate and ammonium sulphate nearly doubled the oil yield from *Mentha piperata* in comparison to that obtained from plants grown without fertilizers. The effect is particularly pronounced when the application is made in the early growth period. Black mints require larger amounts of potash and nitrogen and a relatively smaller supply of phosphorus. Heavy nitrate application and heavy irrigation tend to increase only stem growth and decrease menthol content of the oil. Application of phosphate induces blooming and early maturity.

Interculture—Intercultural operations cover the removal of foreign plants and weeds by hoeing and weeding and keeping the top soil in mulched condition, which checks the rapid evaporation of soil moisture and protects the plants from harmful pests. Weeding and hoeing also facilitate raising of intercrops. In peppermint careful weeding is one of the prerequisites for the production of high quality oil in good yield. As the plants reach the stage of half maturity, hand weeding becomes necessary. The presence of weeds in the herb to be distilled has a very harmful effect upon the odour and flavour of the oil and makes it unsaleable.

We have observed during the cultivation of vetiver that due to profuse vegetative growth, lodging and disintegration of plants takes place. To avoid this while weeding is in progress, it is advisable to tie up the plants and provide proper support for them. To protect the plantation from heavy winds windbreaks should be used.

Harvesting—It is important that essential oil bearing plants be harvested at the right time. It has been observed in foliage crops like mints that in order to obtain optimum oil yield and menthol content, harvesting should be done when the mint is in full bloom. Late cutting reduces the yield of oil. To avoid the drying of the plant material in the sun, harvesting is started early in the day. The duration of vetiver crop varies from 15 to 18 months in south India. In north India 2 year old wild growth is believed to give the highest yield. The wild growth yields 3 to 4 maunds of rhizomeless, washed and dried roots per acre. By proper cultivation as in Java and Reunion, the yield can be increased to 8 to 10 maunds per acre.

Lemongrass is harvested when 4 to 5 feet high. The crop is collected every 6 to 7 weeks. Palmarosa is harvested when flowering has taken place.

Intercropping—When the plantation is young or harvested, the open space in between the rows of the plants may be utilized for raising crops like maize, sawan (*Echinochloa frumentacea*), kodon (*Paspalum scrobiculatum*), maduwa (*Elusine caracana*), and lobia (*Vigna catjang*). In addition to extra income for the cultivator it will bring the benefit of rotation to the soil and, if leguminous crops are grown, nitrogen fixing bacteria will be activated.

The cultivation of essential oil bearing plants will increase the yield of plant material and the essential oil obtained from it ; help in determining the optimum period of growth for the optimum yield of oil; maintain the purity of the variety; enhance the percentage of desirable constituents and eradicate the insect pests and diseases. The cultivation of essential oil bearing plants is being studied at various research centres in the country, but a few years may elapse before authentic information becomes available. To shorten this period it is suggested that all available information should be collected from countries already cultivating these plants, and Indian research centres engaged in experimental cultivation should, from time to time, issue bulletins giving information about the progress made.

Cultural Studies of Essential Oil Bearing Plants Raised in Jammu & Kashmir

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The climatic conditions prevailing in Jammu & Kashmir are not only suitable for the wild growth of a variety of aromatic plants, but also for the cultivation of important indigenous and exotic plants reputed for their essential oils.

The cultivation of important exotic plants, such as, Japanese mint, English dill, lavender, American wormseed and pennyroyal are described. It has been found that all these plants, if cultured properly, develop essential oils which compare favourably with the oils from plants grown in their usual habitat. *Mentha arvensis* **(Japanese mint) yields 2.1 per cent essential oil and 75 per cent menthol comparing well with the plant grown in Japan.**

The physical constants of the essential oils obtained from plants grown in Jammu & Kashmir are compared with those of the oils prepared in foreign countries.

The State of Jammu & Kashmir possesses a variety of climatic conditions that are suitable for the cultivation of a number of indigenous and exotic essential oil bearing plants. Cultural experiments on some of the important aromatic plants introduced in recent years have revealed that English dill (*Anethum graveolens* Linn.), American wormseed (*Chenopodium ambrosoides* Linn. var. *anthelminticum* Gray), *Eucalyptus viminalis* Labill. and *Eucalyptus citriodora* Hook., lavender (*Lavendula officinalis* Chaise), Japanese mint (*Mentha arvensis* Linn.) and pennyroyal (*Mentha pulegium* Linn. and *Mentha piperita* Linn.) can easily be propagated in different localities in the State.

Anethum graveolens Linn. (Syn. *Peucedanum graveolens* Linn.) Dill[1]

It consists of the ripe, dried fruit of the plant *Anethum graveolens* which is indigenous to South Europe and is extensively cultivated in England, Germany, Rumania and in the Mediterranean countries. It does not grow in India. An allied species *Anethum sowa* Kurz containing 3 to 3.5 per cent of oil, having different chemical constituents and physical properties, is cultivated as winter crop in many parts of India. The seeds of *A. graveolens* were obtained through the courtesy of Prof. W. O. James of Oxford University and were sown broadcast in well prepared beds at Yarikah (7,000 ft.) and at Jammu (900 ft.). About 70 per cent of the seeds germinated in two weeks' time and the plant flowered in July at Yarikah and a crop of fruit, too little for chemical analysis, was harvested in September. The vegetative growth of the plant was, however, stunted at this high altitude. At Jammu also the seeds germinated in about two weeks and flowered by the end of February. The vegetative growth of the plant was much better at Jammu than at Yarikah. The fruits were ready for harvest in April. The dry fruits collected from Jammu on steam distillation gave 2.1 per cent of a pale yellow oil with sp. gr.$^{15^\circ}$, 0.876 ; n^{20°, 1.473 and carvone, 45.9 per cent. B.P. standard prescribes: yield, 2.4 per cent ; sp. gr.$^{15^\circ}$, 0.895-0.910 ; n^{20°, 1.481-1.492 and carvone, 43.63 per cent.

From the data it appears that the local dill oil comes up to the official standards and shows the presence of 45 per cent of carvone.

The dill oil and dill water are commonly incorporated in pharmaceutical preparations such as gripe water used in gastro-intestinal trouble in children. The plant is now being propagated on semi-commercial scale. Studies are under progress on the economic production of the dill fruit under treatment of manures and fertilizers.

Chenopodium ambrosoides Linn. var. *anthelminticum*

Chenopodium oil[2] is an important anthelmintic drug used for the expulsion of roundworms and dwarf tapeworms from human beings and live-stock. The oil is obtained from the fresh flowering and fruiting plants of *Chenopodium ambrosoides* Linn. var. *anthelminticum* Gray. The plant is indigenous to the southern States of America and Central America but is now being sucessully cultivated in many parts of the world.

There are six to seven species of *Chenopodium* growing wild in different parts of India but none of them yields the therapeutically active oil. In view of the importance of the drug, its experimental cultivation was started at Mungapoo in Darjeeling and Bangalore in Mysore State sometime ago. The percentage of oil from the cultivated plants was reported to be 0.17 and 0.24 respectively, and the Indian oil showed only 46 per cent ascaridol as against 65 per cent or more present in the American oil. For this and other reasons its cultivation was discontinued.

Some viable seeds were obtained from Turkey through the courtesy of UNESCO and were sown broadcast at a number of places at Jammu (900 ft.), Srinagar (5,000) and Yarikah (7,000 ft.). The seeds germinated in 10-15 days and bore flowers and fruits in the normal way. The growth of the plants was more vigorous at Jammu than at Yarikah and Srinagar where the plants were rather stunted and could not withstand winter. The plants harvested at the fruiting stage from Yarikah and Srinagar showed 1.16 and 0.82 per cent of essential oil with 8.5 and 7.2 per cent ascaridol respectively The plants raised in Jammu were also harvested at the fruiting stage. They yielded a golden yellow oil with unpleasant odour, and a bitter and burning taste. The analytical results of the oil obtained in two successive years from Jammu are given in Table 1.

TABLE 1—CHARACTERISTICS OF CHENOPODIUM OIL OBTAINED IN FIRST AND SECOND YEAR FROM JAMMU PLANTATION AND B.P.C. AND I.P.L. STANDARDS

	FIRST YEAR	SECOND YEAR	B.P.C. STANDARD	I.P.L. STANDARD
Yield, %	1.2 (from the dried fruits and leaves)	0.75 (from the dried fruits and leaves)	0.6 to 1.0 (from the dried fruits)	. .
Sp. gr.	0.9835 at 33°	1.9680 at 33°	0.965 to 0.977 at 15°	0.90 to 0.94 at 15°
n	1.485 at 33°	1.4705 at 33°	1.474 to 1.479 at 20°	. .
Solubility in vol. alc., %	. .	7 to 8 of 70	3 to 7 of 70	. .
*Ascaridol, %	13.4	69.5	Not less than 65	Not less than 40

* The B.P. 1948 method of Cocking and Hyman was employed for the determination of ascaridol.

The oil obtained from the locally grown plants in the second year of growth was also tested for its anthelmintic properties on earthworms according to Sollman's method[2] and it was observed that the oil possessed anthelmintic activity comparable to that of the American chenopodium oil.

In view of the encouraging preliminary results a more exhaustive study is being made on the experimental cultivation and extraction of oil from this plant in Jammu.

EUCALYPTUS SPECIES

Eucalyptus trees are not indigenous to India but many species have been introduced from Australia in different parts of the country notably in Nilgiris in South India and oil of medicinal quality is being distilled there. Attempts to raise eucalyptus were also made in northern India and *E. viminalis* Labill. and *E. citriodora* Hook. were planted in Jammu many years ago. These have grown well under the local climatic conditions and have attained their normal height. The fresh leaves from *E. viminalis* on steam distillation gave 1.4 per cent of a pale yellow oil with sp. gr.$^{15^\circ}$, 0.9007 ; n^{20°, 1.476 ; acid val., 0.6 ; ester val., 16.0 ; cineol 17 per cent ; and soluble in 2-3 volumes of 80 per cent and all volumes of 90 per cent alcohol: values for Australian oil being—yield, 0.35-0.74 per cent ; sp. gr.$^{15^\circ}$, 0.94-0.9162 ; n^{20°, 1.4855 ; sap. val., 9.5 ; cineol, 50 per cent, and soluble in equal volume of 80 per cent alcohol.

The fresh leaves of *E. citriodora* when steam distilled yielded 0.83 per cent of a pale yellow oil with sp. gr.$^{15^\circ}$, 0.9207 ; n^{30°, 1.4924 ; sap. val., 8.5 ; and soluble in 7-10 volumes of 80 per cent and 1-3 volumes of 90 per cent alcohol: values for Australian oil being—yield, 0.596 per cent ; sp. gr.$^{15^\circ}$, 0.864 ; n^{30°, 1.4651-1.4678 ; sap. val., 7.5 and soluble in 1.5 volumes of 70 per cent alcohol.

Lavendula officinalis Chaix

Quite a large amount of lavender oil is used in India in perfumery and soap industries and the whole quantity worth about a lakh of rupees is annually imported from foreign countries. Lavender is indigenous to Europe ; it has been introduced as an ornamental garden plant in India, but no attempt has been made to grow it on commercial scale for the distillation of lavender oil. In France, about 100 tons of oil are produced annually and exported all over the world.

With a view to introduce this plant in Kashmir where climatic and soil conditions are suitable for its healthy growth, viable seeds were procured from France and the seedlings raised and transplanted. The germination was poor in the first instance, but later on the propagation was done by shoot cuttings which did well. The flowering shoots were harvested and dried. On steam distillation they yielded 2.4 per cent of an essential oil having sp. gr.$^{15°}$, 0.9192 and esters, as linalyl acetate, 24.8 per cent ; B.P. requires sp. gr.$^{15°}$, 0.883-0.895 and esters, as linalyl acetate, not less than 35.0 per cent.

Apparently the yield and the quality of the oil obtained from the drug raised in Kashmir is very encouraging and holds out a bright prospect for large scale cultivation of this plant.

Mentha arvensis Linn. (Japanese Mint)

English oil of peppermint is obtained by steam distillation of the fresh flowering tops of *Mentha piperita* Linn. This grows in Europe and is now cultivated in England, America, Russia and many parts of the world. Japanese and Chinese peppermint oils are obtained from *Mentha arvensis* DC. var. *piperascens* Holms and var. *glabra* Holms and are the richest of all oils in menthol.

There are a number of species of mentha which either grow wild or are cultivated in India but none yields the peppermint oil of medicinal value ; large quantities are annually imported. The necessity for the cultivation of *M. piperita* was felt in India as early as 1881 and the plant was raised in the Nilgiris and in Mysore. Recently it has also been raised in Forest Research Institute, Dehra Dun, but the essential oil obtained from these plants did not come up to the standard. The plant has also been raised at a number of places in Kashmir and after repeated trials over a number of years the oil now obtained from the local plants has shown nearly 46 per cent of menthol content. *Mentha arvensis* also grows wild to a limited extent in Kashmir but the oil obtained from these plants does not come up to the official standards and no menthol could be obtained from the oil on chilling.

Recently rooted suckers of *Mentha arvensis* var. *piperascens* were procured through the good offices of UNESCO from Japan and propagated at Jammu (900 ft.) and Srinagar (5,000 ft.). The plants were propagated in Jammu late in March and in Srinagar in early May. The plants in Jammu showed very vigorous growth and bore flowers and fruits in the month of July. In Srinagar, the growth of the plants was rather stunted and did not bear flowers till late in September.

TABLE 2—CHARACTERISTICS OF ESSENTIAL OIL FROM MENTHA ARVENSIS VAR. PIPERASCENS CULTIVATED, AND WILD PLANTS IN KASHMIR, ALONG WITH CHARACTERISTICS OF JAPANESE OIL

	Jammu plants	Srinagar plants	Wild plants	Japanese oil
Yield (from dry leaves), %	2.1	1.6	0.45	1.07 to 1.8
Sp. gr.$^{15°}$	0.92	0.9609	0.9161	0.900-0.912
$n^{20°}$	1.4568	1.4553	1.474	1.4600-1.4635
Solubility in vol. alc., %	7 to 8	6 to 7 of 70	..	..
Congealing point, °C.	14-15.5	16.18	..	16-28
Acid val.	0.64	..	..	0.5-2.5
Menthol acetate, %	13.7	..	..	3.8
Free menthol, %	70.1	..	..	70-90 (total menthol)

The plants were harvested in their flowering and fruiting stage from both the places and the oil obtained was analysed. The quantity of the leaves obtained from plants raised in Srinagar was too small to yield sufficient amount of oil for the determination of the percentage of menthol or menthyl acetates in it. The analytical results of the oil obtained from the leaves of plants grown in Jammu and Srinagar and wild growing plants along with the characteristics of Japanese oil are given in Table 2.

A perusal of the above table shows that the plants raised in Jammu (900 ft.) yield very good percentage of oil. It contains nearly 70 per cent of menthol and compares favourably with the Japanese oil. A large scale propagation of Japanese mint is being attempted at a number of places.

Mentha pulegium Linn. (Pennyroyal)

Pennyroyal oil is used in considerable quantities in perfumery and soap industry. The plant is not indigenous to Kashmir but has been introduced successfully in experimental cultivation. The principal constituent of the pennyroyal oil is a ketone called pulegone. This can be converted into menthone and then to menthol, which is in great demand in India. The dried leaves and flowering tops of the plants on steam distillation gave 2.3 per cent of an essential oil having sp. gr. 15°, 0.8925 and $n^{20°}$, 1.483 ; the Mediterranean pennyroyal oil[4] has sp. gr.$^{15°}$, 0.93-0.95, and $n^{20°}$, 1,483-1.486.

M. piperita Linn.

M. piperita is cultivated on an extensive scale in America, Germany, and England. The plant was cultivated in the Nilgiris in 1818 and has been raised in Mysore and also at the Forest Research Institute, Dehra Dun. Rooted suckers of this plant procured from the Agricultural College, Lyallpur were propagated at Baramula (5,500 ft.), Srinagar (5,000 ft.) and Yarikah (7,000 ft.).

The flowering tops and leaves collected from these nurseries in August were steam distilled. The dry leaves from Yarikah and Baramula yielded 0.7-1.0 and 0.7 per cent oil, respectively. The oil has sp. gr.^{15}, 0.9187 ; *n*, 1.466 ; menthyl acetate, 14.4 per cent, menthol, 46.6 per cent and is soluble in 6 volumes of 70 per cent alcohol. B.P.C. requires: yield, not less than 0.5 per cent ; sp. gr.$^{15°}$, 0.897-0.910 ; *n*, 1.466-1.470 ; methyl acetate, 4.9 per cent ; menthol not less than 46 per cent, and solubility in 4 volumes of 70 per cent alcohol.

Recently rooted suckers of *Mentha piperita* var. *vulgaris* and *M. piperita* var. *officinal* from the U.K. and *M. piperita* from the U.S.A. have been introduced in Kashmir, the plants are being raised in nurseries.

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Rosha Grass Farming as a Cottage Industry

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The author's experiences in cultivation and distillation of rosha grass grown in Jaranwala (West Punjab) before partition and in Bibiwala (U.P.) after partition have been described and a plea for developing rosha grass industry as a cottage industry has been made.

The *motia* variety of rosha grass (*Cymbopogon martini* Wats. syn. *Andropogon martini* Roxb., *A. shoeananthus* var. *martini* Hook. f.) is the best source of palmarosa or rosha grass oil. It is a perennial herb, 5-8 feet high, occurring in the drier localities of Kashmir, Punjab, Bihar, Rajasthan, Madhya Pradesh, Bombay and South India. India which once was the chief exporter of palmarosa oil is now being ousted from her position by Java, mainly because no scientific plantation of rosha grass has been taken up in the country.

Prof. Puran Singh started a rosha grass plantation in 1924 near Jaranwala (West Punjab), which after his death was managed first by Shri J. C. Luthra and then by Dr. C. Khudadad. Dr. Khudadad put the farm in the charge of S. Nirlep Singh, who, in 1938, was succeeded by the author. During the ten years prior to partition of the country, the Jaranwala Plantation produced nearly, 30,000 lb. of "Jaranwala Palmarosa oil" highly valued for its fragrance and its high geraniol content (97 per cent).

The plantation was situated on loamy, well-drained dry soil. Before sowing the seeds in March-April in drills 9 inches apart, the land was ploughed 4-5 times and watered. Great attention was paid to the hoeing and weeding of the crop. The crop was harvested in October and November ; if it was delayed, the frost reduced the oil content.

Steam distillation was carried out at a pressure of 60 lb./sq. in. in a battery of stills working four charges of 5 maunds of grass daily. The yield of the clear light yellow coloured oil after filtration was on the average 1.25 lb. per charge. When coal was not available, steam was generated in an open fire distillation plant by heating water below the false bottom of the still on top of which the charge of grass was placed. This required very careful handling, as the slightest charring of grass could spoil the quality of the oil. In this method the exhausted grass can be used as fuel. The yield of oil is lower but its quality is superior due to the lower pressure of the steam which does not allow the higher boiling pungent aldehydes to distil over. The average yield of the oil in Jaranwala farm was 15 lb. per acre.

A rosha grass plantation was started by the author in India (100 acres granted by the Government of U.P.) in Bibiwala Forest Blank in 1952. Seeds obtained from Madhya Pradesh Forest Office were used. It was discovered that the best time to sow rosha grass in weed infested area is August after thorough weeding and ploughing. The heavy rainfall of the locality was conducive to good vegetative growth and the plants attained a height of 8-11 ft. with broad leaves. The flowers and the seeds, however, were not so well developed as in the Jaranwala plantation. As pure seeds of *motia* variety were not available, the plantation, in addition to this variety, contained four other varieties and lot of care was necessary to eliminate them. The propagation of rosha grass by root cuttings and stem nodules is possible, but this process, though capable of yielding pure *motia* plantation, is expensive.

The yield of oil produced in an open fire distillation plant was the same as at Jaranwala but it had a better odour value, possibly due to the higher altitude of Bibiwala. By collecting the earlier distilling fractions separately, the geraniol content of the oil distilled in November was raised to 96 per cent. The oil had a ready foreign market and fetched a high price.

The following conclusions drawn from the experience of the author are worth recording:

1. Rosha farming on a cottage industry scale has bright prospects in U.P., where barren forest lands could be utilized.
2. By spending about Rs. 600 per acre, it is possible to raise a plantation which will yield 30 lb. of oil per year for several years.
3. Purifying the oil and isolation of pure geraniol from it may bring better returns.
4. Rose cultivation along with rosha grass cultivation, if adopted, may help stabilize the industry as rosha grass oil can be used for mixing with rose oil as there is a ready market for the mixed oil.

Lemongrass Cultivation in Travancore-Cochin State

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The details of lemongrass cultivation in Travancore-Cochin are given and the work done for the development of lemongrass oil industry at the Lemongrass Research Institute at Perumbavoor is discussed.

Among the more important essential oils produced in India, lemongrass oil, which is the oil obtained by the distillation of the aromatic grass *Cymbopogon flexuosus* Stapf. occupies probably the second place only to Mysore sandalwood oil. In contrast to the sandalwood oil, which is produced on a factory scale using modern techniques, lemongrass oil is even now extracted by a crude and primitive method. It is reported that a scheme for the cultivation and distillation of lemongrass on scientific lines was initiated in Malabar by an American concern. The cost of the distillation plant erected was reported to be Rs. 12,000 and the extraction of oil was carried out on a factory scale. The scheme has been wound up. It was one of the very few attempts made to produce lemongrass oil on a factory scale in this country.

The main centres of cultivation of lemongrass are confined to Travancore-Cochin State and certain parts of Malabar where it exists both in the cultivated and wild forms. The estimated area under cutivation in the Travancore-Cochin State is about 36,500 acres. Quite recently, attempts have been reported to be made to cultivate lemongrass in Assam under a scheme sponsored by the Union Government as an experimental measure with the seeds of the red variety of grass sent to Assam.

Cultivation—The cultivation of the grass on extensive scale was started in Travancore-Cochin only about 60 years ago. It flourishes in well-drained sandy loam and cannot withstand water-logging. As a rule, it is cultivated in poor soils not fit for further improvement. Vast stretches of such land are under this grass in the northern talukas of the State. The warm and humid climate of the region alternating between sunshine and showers is very conducive to its good growth. It requires an annual rainfall of about 100 inches and is cultivated up to an altitude of 4,000 to 5,000 ft.

The sowing season begins in March-April depending on the incidence of early rains of the South-West monsoon. Seedlings are raised in a finely prepared nursery just after the first showers and later transplanted. One maund (82 lb.) of seed is sufficient for transplanting more than 3 acres of land. The seeds under favourable conditions germinate in 4-5 days and the seedlings are ready for transplanting after about 50-60 days. Transplanting is done along raised beds about 3 ft. wide at a distance of 4-5 in., the different ridges being separated by furrows. Transplanting commences by the middle of June and the plants are ready for the first cutting about 90 days after. Immediately after the first cutting one weeding is given but earthing up is done only during the second and subsequent years. Normally no manures are applied by ordinary cultivators.

The second and subsequent harvests are taken at intervals of 40-45 days depending on the fertility of soil and seasonal factors. In the first year of planting it is generally possible to harvest the grass 3 or 4 times while in subsequent years the number of cuttings may be 4 or 5. The harvesting is done with the aid of sickles ; the clump is cut closely at a height of 3-4 inches above the ground and tied into small bundles. About 15 women are required to harvest one acre of land. The grass thus harvested is then carried for distillation to a shed close by. The operations come to a close by the end of December. During the summer, the leaves dry and shrive, but the roots survive. In certain localities there is the practice of setting fire to the grass in summer. After the first rains the old roots sprout, the plant again begins to grow and puts forth new foliage which is harvested and distillated. Normally a plantation lasts about 5 years, but in better soils the life of a plantation might extend up to 8 years. In rare cases the crop survives for a period of about 12-15 years. Replanting becomes necessary when the stand becomes very poor and the yield is uneconomical.

In forest regions where extensive areas are brought under cultivation direct broadcasting of seeds is practised. Due to thick stand in such plantations weeding becomes difficult and their life is reported to be considerably shortened.

Lemongrass though generally propagated from seeds can also be raised by division of clumps or clonal multiplication. As plants subjected to regular harvesting will not yield seeds, separate plots are maintained for the production of seeds. Seeds are collected during the months of December and January.

Yield of oil—The yield of oil is the higest during the second and third years after planting. The best season for harvesting is from September to December when the yield and the citral content are the maximum ; they are reduced if the grass is cut during heavy rains. The yield of freshly harvested grass from an acre is about 15,000 lb. per year ; the yield of oil being about 35-40 lb. in the first year, and 45-50 lb. during the second and third years. The actual yield obtained at this Research Station in 1953 (second year of planting) from one acre of plantation is given in Table 1. It is interesting to note that the yield of fresh grass, oil and the citral percentage vary from month to month. It may be mentioned in this connection that Ganesh Chandra and G. N. Gupta tried experimental cultivation of lemongrass in Uttar Pradesh and obtained a yield of 0.14 to 0.22 per cent (on the weight of fresh grass) and citral content of 70-75 per cent.

Varieties—There are several varieties of lemongrass in Travancore-Cochin. Two of them, the red lemongrass with its red stem and the white lemongrass with its white stem are popularly recognized. The oil obtained from the red variety is of superior quality and is the genuine lemongrass oil. The oil from the white variety is of an inferior quality with very low citral content and is insoluble in 70 per cent alcohol.

Characteristics of oils from both varieties of the grass were determined by Varier[2] and the results are given in Table 2. It is evident that although the white grass gives about 50 per cent higher yield of oil, the oil contains very little citral and is therefore useless from the commercial point of view. As the oil is used for adulteration of genuine oil it is advisable that the white grass should be exterminated.

Citral—Before purchasing a bulk of oil the merchants test the sample with regard to its citral content, as the value of the oil is mainly determined by the percentage of citral present in it. Citral is the major constituent of pure lemongrass oil and is industrially used in the preparation of ionone, an important ingredient of perfumes and cosmetics. The price of the oil is fixed in terms of citral per cent and the minimum percentage acceptable in trade is 77. The citral content of the oil extracted in the northern districts of the State is much higher than the minimum required and hence the practice is to adjust the citral content at 77 per cent by mixing oils low in citral content with citral-rich oils.

The development of citral in the plant is influenced by both edaphic and

TABLE 1—YIELDS OF LEMONGRASS AND LEMONGRASS OIL AND CITRAL CONTENT OF THE OIL*

HARVEST NO.	DATE	INTERVALS *days*	YIELD PER ACRE			CITRAL %
			Fresh grass *lb.*	Oil *lb.*	Oil %	
1	7-5-1953	. .	1,772	9.4	0.53	79
2	24-6-1953	47	2.624	11.7	0.44	84
3	10-8-1953	46	3,950	11.8	0.30	84
4	22-9-1953	42	2,940	10.5	0.35	80
5	4-11-1953	42	3,450	12.2	0.35	83
6	14-12-1953	39	1,825	9.0	0.49	81
	Total		16,561	64.6		

* At different harvests from a plantation at Perambavoor Lemongrass Research Station (1953)

TABLE 2—CHARACTERISTICS OF OILS FROM RED AND WHITE VARIETIES OF LEMONGRASS

	RED GRASS OIL	WHITE GRASS OIL
Yield, %	1.1	1.7
sp. gr.$^{30^\circ}$	0.881	0.931
n^{30°	1.482	1.498
Aldehydes as citral, %	76.4	8.9
Solubility in 70% alc., vol.	2.8	Insoluble

TABLE 3—YIELD OF OIL AND ITS CITRAL CONTENT OBTAINED FROM DIFFERENT TYPES OF LEMONGRASS (1953)

No.	Types	Yield of oil from 10 lb. cc.	Citral %
1	Mannangandam	4.8	85.84
2	Shengulam	4.7	86.52
3	Kallayi	6.0	65.34
4	Dehra Dun	10.0	64.80
5	Ceylon	14.1	11.37
6	Pattikkad	3.8	83.97
7	Nemmara	7.2	76.10
8	Vengoor	6.0	85.65
9	Kavalangad	10.0	82.85
10	Vadakkancherry A	10.0	73.02
11	Vadakkancherry B	6.4	75.80
12	Kulathupuzha	10.0	76.20
13	Konni Estate	8.8	75.84
14	Chelad	7.1	84.41
15	Valayanchirangara	8.5	86.12
16	Konni	12.5	87.62
17	Local	7.5	81.40

climatic factors. The citral content of the same plant in different seasons and at different stages of growth is subject to variation. The influence of the application of fertilizers and of harvesting at different periods of growth in the plants ontogeny on the yield of oil and the amount of citral content is under investigation at the Lemongrass Research Station.

Improved varieties—For the development of lemongrass oil industry it is essential that better varieties of grass should be cultivated. With the object of producing superior varieties of lemongrass by selection and hybridization work on a scheme of research at the Lemongrass Research Station is in progress. The possibilities of evolving a new variety with not only a higher yield of oil but also with a higher citral content are being explored. A fairly large collection of lemongrass from different parts of the State has been made and is being studied. Some of the selections were tested with regard to yield and citral content on a preliminary scale and the results are furnished in Table 3. It may be noted that the performance of one particular culture (Type 16) appears to be very good with regard to both the yield of oil and the citral content.

By-products—The waste grass obtained after the extraction of oil may be used as fuel, fodder and compost manure.

Pests and diseases—Lemongrass plants are not generally infested by any disease or pest.

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Production of Alcohol of Perfumery Grade

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The impurities, particularly aldehydes, contained in commercial rectified spirit make it unsuitable for perfumery industry and also for the preparation of alcoholic caustic potash used in oil analyses. Methods based on absorption, oxidation and reduction have been tried for the removal of these impurities. A reduction method employing sodium amalgam has been developed and found to yield satisfactory results.

Rectified spirit finds extensive use as solvent and diluent in the manufacture of perfumes and flavouring agents. Due to import restrictions imposed by the Government of India, Indian perfumers are experiencing great difficulty in obtaining alcohol of requisite quality. The rectified spirit manufactured by Indian distilleries contains various odorous impurities which adversely affect the quality of the perfumes. Even the specifications laid down by the Indian Standards Institution for rectified spirits allow for traces of impurities like aldehydes, acids and higher alcohols[1]. The rectified spirit required by the perfumery industry is almost entirely free of aldehydes and higher alcohols, and is termed 'Extra Neutral Alcohol' or 'Silent Spirit'.

The 'silent spirit' is commonly manufactured by distilleries in France and the United States of America. In an attempt to prepare alcohol suitable for the perfumery industry, the authors have tried chemical methods for the purification of industrial alcohol.

As aldehydes and higher alcohols, particularly the aldehydes, are mostly responsible for making alcohol unsuitable to the perfumery industry and also for the preparation of colourless alcoholic caustic potash solutions used in volumetric analysis, attempts were concentrated on the removal of these impurities. The methods employed may be broadly calssified into four groups: (*i*) those involving use of absorbent charcoals and earths (*ii*) oxidation (*iii*) reduction and (*iv*) methods in which impurities are fixed by formation of complex compounds with the reagents used.

Absorption methods

A method based on the technique employed by Zaharia[2] was tried. Four types of active charcoal, namely, wood, blood, Norit and Pick, and fuller's earth were investigated for their efficiency in removing impurities like acetaldehyde, furfural, acetic acid and fusel oil. It was noticed that the absorption of impurities is directly proportional to the concentration. Higher alcohols and fusel oil were the most easily absorbed. They are almost completely removed on treatment with wood charcoal. Aldehydes and acids were the most difficult to remove. It was not found possible to eliminate last traces of aldehydes even after repeated treatment.

Among the charcoals, wood charcoal was found to be the most effective. Fuller's earth was found to be much superior to any charcoal for removing acids and higher alcohols, although aldehydes were not completely removed. Experiments in which the original alcohol contained 0.1 g. of acetic acid and isoamyl alcohol per litre showed that after filtration through a 12 in. column of fuller's earth at the rate of 5 ml. per minute, the amount of acid was reduced to 1 part in 100,000 parts of alcohol and of isoamyl alcohol to 3 parts in 100,000.

The above methods have a major drawback in that the absorbent has to be repeatedly regenerated and reactivated. The loss of alcohol by absorption is also appreciable unless large quantities are treated.

Oxidation methods

Alkaline potassium permanganate appears to be the most suitable oxidizing agent for this purpose. The treatment oxidizes the aldehydes to acids and fixes them as alkali salts. On distillation, the reaction mixture yields a purer sample of alcohol. Sodium peroxide can replace potassium permanganate in the reaction. As during distillation some alcohol is again oxidized into aldehyde, it was not possible to prepare alcohol absolutely free from this impurity. Distillation at reduced pressure helps in keeping the quantity of aldehydes low but it is again formed on standing. Alcoholic caustic potash, prepared from alcohol purified in this manner, developed a faint yellow colour after 4 days.

Complex formation methods

For the production of aldehyde-free ethyl alcohol in the laboratory, treatment with *m*-phenylene diamine hydrochloride has been found to be successful[3]. Samples of alcohol which contained 0.5 per cent acetaldehyde were refluxed for 48 hr. with *m*-phenylene diamine hydrochloride, and the product distilled. This treatment reduced the aldehyde content to 1 part in 100,000 parts of alcohol. *m*-Phenylene diamine hydrochloride is rather expensive. If the impure sample of alcohol is digested with caustic alkali to remove a major portion of the aldehydes by resinification, a smaller amount of *m*-phenylene diamine is consumed.

Reduction methods

These involve the treatment of the impure alcohol with reducing agents which convert the acetaldehyde back into ethyl alcohol. The reducing agents employed by Sakaguti[4] were reduced nickel and copper. Castille and Henri[5], and Louis Harris[6] have suggested the use of granular zinc and zinc wool (Kahlbaum) respectively. Sodium hydrosulphite has been suggested by Englis and Mills[7], whereas Winkler[8] has found silver nitrate useful.

In reduction treatment, the strength of the final product goes down, and it is advisable to start with very high strength rectified spirit if the desired product is to be 90 per cent alcohol.

TABLE 1—SAPONIFICATION VALUES OF OILS USING CAUSTIC POTASH SOLUTIONS OBTAINED BY USING ALCOHOL PURIFIED BY DIFFERENT METHODS

OIL	SILVER OXIDE	STOUT & SCHUETTE METHOD	MALFATTI METHOD	SODIUM AMALGAM METHOD
Sesame	190.2	190.8	188.8	190.0
Cottonseed	194.4	194.8	194.4	194.4
Coconut	256.3	256.7	256.1	256.3
Mahua	190.0	190.4	188.6	190.4

To a certain extent, this drawback is eliminated by the method of Stout and Schuette[9] who use granular aluminium in the presence of alkali as the reducing agent. For one litre of alcohol, 8-10 g. of caustic potash and 5-10 g. of granular aluminium are used.

In the present work, the authors have used sodium amalgam for preparing an anhydrous aldehyde-free product. In 8 out of 10 experiments it was possible to get absolute alcohol with aldehyde content reduced to almost zero. The success of purification is indicated by the fact that when 10 ml. of the final product were mixed with 10 ml. of dilute potassium permanganate, it took more than one hour for potassium permanganate to be decolourized. The alcohol, when stored in dark bottles, remained aldehyde-free for 4 months. The product when tested as a solvent for perfumes showed encouraging results. Alcoholic caustic potash solutions prepared from the alcohol were in no way inferior to those obtained from the alcohols purified by the methods of Stout and Schuette[9] or Malfatti[10]. The solution remained almost colourless for 2 months before developing a very faint yellow coloration. The alcohol purified by the Stout and Schuette method under identical conditions developed coloration after 3 weeks and the caustic potash solution prepared with the alcohol treated by Malfatti process became coloured after 33 days. The alcoholic caustic potash solution was used for the determination of saponification values of cottonseed, coconut, *mahua* and *til* oils, and was found to be quite satisfactory as indicated in Table 1.

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Studies on Extraction of Citrus Oils

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A systematic investigation of the different methods of extracting citrus oils from fresh and dried peels of oranges and limes, and flowers of Coorg orange has been carried out.

The esssential oils of citrus peels, flowers and leaves have considerable economic importance. They are used as flavouring agents in beverages, baked goods and pharmaceuticals and are widely employed in the preparation of perfumes and cosmetics. Although India is rich in these raw materials, fairly large quantities of these oils are imported from countries like Italy, Sicily and Palestine. Considerable work has already been carried out in Italy, Sicily, Palestine and other parts of the world on the extraction of citrus oils[1-3]. Recently, Guenther[4] has given a detailed account of the methods of their production. In India, however, no systematic work has been done on the recovery of these oils. Saletore[5] and Patel[6] have outlined some of the methods of extraction and Rao *et al.*[7] and Nigam & Dutt[8] have given data on their physico-chemical characteristics. In view of the importance of citrus fruits in the indigenous fruit preservation industry, it was felt necessary to work out methods for the profitable recovery of oil from the peels and also to find out ways and means of making use of leaves, cuttings, and flowers, which are likely to be available in increasing quantities, for the extraction of oil. Detailed investigations were, therefore, undertaken under the auspices of the Essential Oils Research Committee of the Council of Scientific & Industrial Research. Some of the results of these investigations are briefly discussed in this paper.

Material and methods

The materials used consisted of fresh as well as dried Coorg orange peel, dried Nagpur orange peel, fresh *Sathgudi* and *Musambi* orange peels, lime peel, sweet orange leaves and flowers, bitter orange and pummelo flowers. For the recovery of oil, cold extraction as well as distillation methods were used with several modifications. The effect of preliminary treatment of the orange peels such as mincing, steeping in lime water, and soaking in calcium chloride solution, was also studied.

Cold expression of oil from whole loose jacket oranges

In the case of whole oranges, in addition to the well-known method of scraping the oil from the epidermis of the peel with a spoon, a potato peeler which has a revolving abrasive disc was used to grate the oil cells in the peel ; and, subsequently, the oil in the washings of the scrapings was recovered by steam distillation. The yield of oil, using one hundred oranges, was almost nil. Oranges were badly bruised, although limes stood the test fairly well. A large rotary copper drum with its inner surface punctured to give pyramidal projections was also tried to abrade the surface of the peel. The results were not quite satisfactory. The yield of oil from hundred Coorg oranges of average size was 5 to 6 ml. only. A pin-mat which consisted essentially of a circular plate of paraffin wax studded with sharp pointed pins, projecting just sufficient to lacerate the oil-bearing epidermis of the peel without injuring the segments inside, was also tried. The oil from the cell sap, which was collected by rolling the whole orange on this sharp pin-mat, was subsequently separated by centrifuging. The yield of oil was about 5 to 6 ml. from 100 Coorg oranges weighing 20 to 25 lb. The defect in this method was that, in spite of precautions, the fruit was punctured deep in some parts and the treated oranges did not keep even for 2 to 3 days. Dipping them in oil or coating them with wax did not help in prolonging their keeping quality. The oranges treated by the pin-mat method have, therefore, to be utilized without much delay for the extraction of juice or conversion to other products which can be preserved suitably.

Cold extraction of oil from orange peels

Several methods were tried to extract the oil from the fresh peels with varying degrees of success.

Hand pressing—The fresh orange peel was squeezed by bending it between the thumb, index finger and the middle finger so that the oil cells in the outer epidermis of the peel burst open and the oil squirted out with some force. The spray was directed towards a glass funnel placed on top of a glass bottle so that the oil drops collected gradually in the bottle. Any liquid sticking to the peel was scraped against the sharp edge of the funnel. The yield of oil was about 2 ml. per lb. of peel and it took nearly an hour to get it. Although the process was slow, the oil obtained was of excellent quality, approximating to that present originally in the oil glands of the peel.

Other methods—Other methods, such as (*i*) mincing the peel and subjecting it to high pressure (*ii*) maceration of the minced peel with water and (*iii*) crushing the peel between two granite rollers, yielded emulsions which were difficult to break and separate into distinct oil and sap layers. Crushing the rolled peel between two metal plates by applying pressure by means of a hand lever also did not lead to satisfactory extraction of the oil.

Extraction in a roller mill—A gun metal two-roller press fabricated in the laboratory was tried next. This consists essentially of two adjustable rollers revolving towards each other when turned by means of a handle. The top roller has 1/16 inch grooves made in such a way that small projecting squares

TABLE 1—EFFECT OF PRELIMINARY TREATMENT OF PEEL ON YIELD OF OIL

PEEL TO WATER RATIO	SOAKING TIME *hr.*	STRENGTH OF LIME WATER %	YIELD OF OIL		
			MIN. %	MAX. %	AV. %
1:3	4	0.075	0.50	1.40	0.89
		0.10	0.64	1.10	0.89
		0.25	0.37	1.19	0.91
		0.50	0.60	1.06	0.86
1:6	4	0.10	0.74	1.17	1.02
		0.25	0.90	1.00	0.90
1:6	2	0.10	0.70	0.82	0.65
		0.25	0.50	0.72	0.62
1:3	8	0.10	0.80	0.94	0.88
Nil	Nil	Nil	0.10	0.30	0.20

TABLE 2—CHARACTERISTICS OF OILS FROM TREATED (CaO, 0.075-0.5 PER CENT) AND UNTREATED FRESH ORANGE PEELS

	OIL FROM TREATED PEEL			OIL FROM UNTREATED PEEL		
	Min.	Max.	Av.	Min.	Max.	Av.
Sp. gr.	0.8479	0.8488	0.8483	0.8483	0.8485	0.8484
n	1.4746	1.4750	1.4746	1.4748	1.4748	1.4748
$[\alpha]_D$	+100.02°	+103.16°	+100.36°	+100.17°	+101.67°	+101.17°
Evaporation residue, %	5.09	5.92	6.05	5.57	5.88	5.76
Acid val.	1.57	1.91	1.87	1.35	1.80	1.65
Ester contents, %	3.06	3.92	3.23	2.94	3.92	3.27

of 3/16 inch side are formed on its surface. The bottom roller has circular grooves cut 1 mm. wide so that sharp ridges are formed on it. When the orange peel is placed between these two rollers with the white inside portion of the peel against the squares in the top roller and the oil-bearing outer surface of the peel against the narrow cutting ridges of the bottom roller and the handle turned, the peel is drawn in between the rollers and the oil, together with the sap, is pressed out. This emulsion of oil and sap is collected in a tray placed below. The expressed oil does not come into contact with the spongy albedo portion of the peel. This is decidedly a big advantage because, once the albedo portion absorbs the expressed oil, it is almost impossible to press it out again in the cold. The use of this roller press having given very encouraging results, it was used in a series of trials.

About 50 batches of fresh peel of Coorg orange were pressed in this roller press. It was, however, noticed that for satisfactory working, it was necessary to harden the peels to some extent. Lime water was found to be suitable for this purpose. The effect of the ratio of peel to water and of the concentration of calcium oxide on the yield of oil is recorded in Table 1. The oil on the top layer was obtained by separating this layer with the help of a spoon and squeezing it in a piece of thick flannel. It will be seen that the maximum yield of about 1.2 per cent of oil can be obtained by soaking the peel for 4 hr. in 0.1 to 0.25 per cent lime water. Although the increase of the ratio of peel to water from 1:3 to 1:6 did not have any effect, it is desirable to adopt the latter ratio to cover the peels satisfactorily. Increase of the strength of lime water much beyond 0.25 per cent renders the peel highly brittle and unsuitable for extraction. The very low yield of 0.1 to 0.3 per cent of oil in the case of peels that were not steeped in lime water shows the advantage of lime treatment.

Analysis of the peel oil—Samples of oil obtained by different treatments of the peel were analysed for their physico-chemical constants. The results are shown in Table 2.

It will be seen that the values for the oil from the treated peel fall within the range of those obtained for the oil from untreated peel.

Cold expression of oil from dried orange peel

The use of fresh orange peel for the extraction of oil has a limitation in that the peels have to be used almost immediately ; which is not always possible. To overcome this difficulty, the possibility of extraction of oil from the dried orange peels was investigated in detail. Soaking for 16 hr. in water gave a low yield of oil ranging from 0.04 to 0.44 per cent only. In 5 out of 18 experiments, the recovery of oil was almost nil. Soaking the peels in ten times their weight of calcium chloride solution containing 0.25, 0.50 and 1.0 per cent calcium chloride showed that the yield of oil was as much as 0.50 to 1.90 per cent, although the peels used had been stored for several months. Decrease in the strength of calcium chloride solution to 0.125 per cent lowered the yield of oil considerably. Soaking in calcium chloride solution thus facilitates the cold extraction of oil from dried peels.

Analysis of the cold-pressed oil of dried orange peel—The physico-chemical constants of cold-pressed oil of dried orange peels are given in Table 3. There was no marked difference in the values for the oils obtained from the peels treated with different concentrations of calcium chloride in the steeping solution.

It will be seen that the values for evaporation residue and free acidity in particular, and for ester content, to some extent, are abnormal, probably due to changes taking place in quality of oil in the peel during storage. Further experiments are, however, needed to determine the relative effect of factors such as the degree of drying and period of storage of the peel on the quality of oil.

Steam distillation of citrus oils

Although the steam-distilled oil is slightly inferior in quality to the cold-pressed oil and fetches a much lower price than the cold-pressed oil, it finds use in cases where the high-priced cold pressed oil cannot be used. Preliminary trials having shown that 26 hr. are required for distillation to exhaust the orange peel of its oil, experiments were carried out to decrease this time. The data for fresh and dried orange peels are given in Tables 4 and 5. The theoretical yields of oil in the peel as determined by Clevenger's method are also shown for comparison.

Mincing of the peel was of great help in facilitating the distillation of the oil from fresh as well as dried orange peels within the first 2-4 hr. It reduces the cost of production to a considerable extent.

Analysis of steam distilled orange peel oils—The averages of physico-chemical characteristics of steam-distilled oils of fresh as well as dried peels of loose jacket mandarin oranges are shown in Table 6.

The earlier fractions of the oil are low in acid value which increases towards the end of the distillation. The quality of the oil is, therefore, likely to deteriorate further when the distillation is prolonged as in the case of the unminced peel. It is, therefore, preferable for steam distillation, to mince the peel.

Sathgudi orange and lime peel oils

Minced fresh *Sathgudi* orange peels, fresh lime peel and fresh lime peel pulp were steam distilled and the recovered oils analysed (Table 7). In the case of *Sathgudi* orange peel, the average yield of oil was 1.8 per cent at the end of 7 hr., the theoretical yield being 2.91 per cent. As in the case of the Coorg orange peel, the first fraction of the oil had higher ester content and less acid value than the end fractions.

Effect of the storage of peel on the yield of oil

Fresh Coorg orange peels were dried in the sun and filled into gunny bags which were stocked in a room. The yield of oil was 1.25 and 0.7 per cent at the end of one and four months' storage respectively ; showing that on prolonged storage the yield of oil decreases considerably.

Essential oil from the leaves of Coorg orange tree

The leaf oil known as 'petitgrain oil' is slightly greenish in colour and finds ready use in perfumery on account of its pronounced aroma. In a trial, using 358 lb. of leaves from Coorg, the yield of oil was 0.14 per cent and the time taken for water-steam distillation was 5½ hr. The oil had the following characteristics: sp. gr., 0.8543 ; n, 1.4702 ; one volume of oil soluble in 5 volumes of 90 per cent alcohol.

Essential oil from sweet orange flowers

The oil from orange flowers known as 'neroli oil' is an expensive and highly prized essential oil used in perfumery. In a trial using 50 lb. of flowers in

TABLE 3—CHARACTERISTICS OF OILS OBTAINED FROM TREATED* AND UNTREATED DRIED ORANGE PEELS

	OIL FROM TREATED			OIL FROM UNTREATED		
	Min.	Max.	Av.	Min.	Max.	Av.
Sp. gr.	0.8477	0.8531	0.8503	0.8526	0.8543	0.8533
n	1.4721	1.4750	1.4735	1.4741	1.4750	1.4747
$[\alpha]_D$	+96.70°	+105.57°	+100.6°	+96.57°	+96.70°	+96.67°
Evaporation residue, %	6.4	13.4	10.1	10.8	12.8	11.7
Acid val.	2.91	7.40	5.21	5.83	7.18	6.36
Ester contents, %	1.96	4.8	3.34	5.23	6.54	6.10

* $CaCl_2$, 0.25-1.0 per cent

TABLE 4—EFFECT OF MINCING FRESH ORANGE PEEL* ON RECOVERY OF OIL BY STEAM DISTILLATION

MATERIAL AND TREATMENT	TIME TAKEN FOR DISTILLATION *hr.*	YIELD OF OIL %	THEORETICAL YIELD OF OIL BY CLEVENGER'S METHOD %
Whole fresh orange peels, not minced, continuous distillation	26	2.4	. .
Whole fresh orange peels, not minced, discontinuous distillation	18-28 (spread over 4 days)	1.3 -2.1	1.90
Fresh orange peel, minced using 11/64 inch plate	4 -5	1.20-1.73*	1.85

* The peel used was rather low in oil content ; 60-65 per cent of the oil distilled over in the first hr. and 35-40 per cent in the next 5 hr.

TABLE 5—EFFECT OF MINCING DRIED ORANGE PEELS ON RECOVERY OF OIL BY STEAM DISTILLATION

MATERIAL AND TREATMENT	TIME TAKEN FOR DISTILLATION *hr.*	YIELD OF OIL %	THEORETICAL YIELD OF OIL BY CLEVENGER'S METHOD %
Whole peels, not minced, continuous distillation	26	1.0 -2.1	2.0
Whole peels, not minced, discontinuous distillation	14½-22½ (spread over 4 days)	0.60-1.73	2.0
Peel minced using 11/64 inch plate	5 - 6½	2.2 -3.0*	3.73
Peel minced using 6/64 inch plate	5 - 6½	3.3 -3.7**	3.73

* 73-78% of the oil was collected in the first 1½ hr.
** 88-90% of the oil was collected in the first 1½ hr.

TABLE 6—CHARACTERISTICS OF STEAM-DISTILLED ORANGE PEEL OILS

	Oil of fresh peel		Oil of dried peel	
	Collected in the first hr.	Collected at the end of 5 hr.	Collected in the first hr.	Collected at the end of 5 hr.
Sp. gr.	0.8398	0.8404	0.8381	0.8402
n	1.4705	1.4705	1.4710	1.4716
Acid val.	0.67	1.57	0.93	4.86
Ester, %	1.30	1.30	1.52	1.47

TABLE 7—CHARACTERISTICS OF SOME STEAM-DISTILLED CITRUS OILS FROM FRESH PEELS

	Sathgudi orange peel oil	Lime peel oil	Lime pulp oil
Theoretical yield of oil (by Clevenger's method), %	2.91	0.17	. .
Oil recovered by distillation of minced material, %	1.80	0.12	0.83
Sp. gr.	0.8399	0.8417	. .
n	1.4713	1.4705	. .
Acid val.	0.55-1.11	. .	. .
Ester content, %	1.30	. .	. .

five water distillations, the yield of oil was 0.08 per cent as against a yield of about 0.10 per cent reported in literature from countries like France, Italy and Haiti. The calyx and other green portions of the flowers have to be removed as they impart a "raw odour" to the oil. The analysis of the oil was: sp. gr., 0.8386-0.8441 ; n, 1.4688-1.4705 ; acid val., 0.75-2.24 and solubility, one volume in 5-9 volumes of 90 per cent alcohol.

On account of the high price it fetches the recovery of neroli oil from dropped flowers is a worthwhile proposition. The main difficulty, however, is that of the collection of the dropped flowers. Mats and corrugated sheets, have to be spread below the trees for collecting them. This is a costly procedure.

Small lots of flowers of the bitter orange and the pummelo were also distilled, but the yields of oils were low.

Conclusions and Summary

Of the various methods studied for the cold extraction of oil from the fresh as well as dried peels of oranges, the use of a specially devised two-roller mill has given satisfactory results as regards the yield and quality of the oil. In this method, the preliminary soaking of the fresh peels for about 4 hr. in 0.10-0.25 per cent lime water and of dried peels for about 16 hr. in 0.5-1.0

per cent calcium chloride solution increases the yield of oil. Treatment with calcium oxide or calcium chloride does not affect markedly, the constants for the oil.

Mincing of the fresh as well as the dried orange peel prior to steam distillation is necessary to facilitate the distillation of the oil within a short period of 4 to 5 hr. as against 20 to 26 hr. required in the case of the whole peel. This also helps in the recovery of oil of better quality since on prolonged distillation the quality of the oil deteriorates considerably.

Steam distillation of *Sathgudi* orange peel and lime peel gave a yield of 1.80 and 0.12 per cent of oil, respectively.

On prolonged storage of dried orange peels, the yield of oil decreases considerably.

Dropped flowers of the Coorg orange tree yield about 0.08 per cent of neroli oil of high quality. The cost of their collection is, however, very high. The fresh leaves of the Coorg orange tree yield on distillation about 0.14 per cent of petitgrain oil of good quality.

The results of these investigations on the recovery of citrus oils are promising. Further large scale trials are, however, necessary to work out the economics of their application on a commercial scale.

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Indian Mandarin Oils : Their Physico-chemical Composition and Future Scope

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The present status and future scope of the citrus oil industry in India with special reference to mandarin oils have been discussed.

The results of studies on typical samples of cold-pressed Nagpur mandarin oils are presented and compared with the available data on other Indian and foreign mandarin oils.

Citrus oils are undoubtedly indispensable in the manufacture of alcoholic and non-alcoholic beverages like mandarin liquors, citrus squashes, syrups, aerated waters, products like citrus marmalades, candies, preserves, baked goods, confectionery, condiments, ice-cream, pharmaceuticals and toilet products[19,23]. There is no organized citrus oil industry in India ; the total annual production of citrus oils in 1951 and 1952 being only 149 lb. and 291 lb. respectively.

In India only two types of citrus oils are produced on a small scale : lime oil is distilled in Utrain in East Khandesh (Bombay State) and mandarin oil in Coorg and Nagpur. The entire demand for citrus oils is, therefore, met by imports. The total imports of citrus oils are presented in Table 1.

Until recently, Italy was the main source of supply of citrus oils, but the United States of America and very recently Palestine and Sicily have succeeded in developing the citrus oil industry considerably[19]. Other principal producers of citrus oils are Brazil, French West Africa, Algeria, Spain and Mexico. Sicily and Spain are the major producers of mandarin oil.

Despite the fact that citrus fruits are among the major fruits grown in India, ranking third in acreage[8], the citrus oil industry has not yet received the attention it deserves. Of the entire citrus production, oranges (both loose as well as tight jacket) alone constitute about 79 per cent[7]. The loose jacket oranges (mandarins) are available in fair abundance in Madhya Pradhesh, Bombay, Coorg, Assam, Sikkim, Madras, Punjab and Pepsu and at present, a considerable quantity of the fruit goes to waste because of lack of transport and marketing facilities. Besides, a fair amount of unripe fruits fall off every season due to winds, hailstorms and go to waste. In addition, there is a considerable amount of 'cannery refuse' from the various factories all over the country, which mostly go unutilized. If an attempt were made to recover oil from the peels of waste fruit and from cannery refuse, considerable income would accrue both to the planters and to the preservers. Mandarin peel oil, like other citrus oils, is equally important and because of the diverse uses to which this oil can be put, it is hoped that it will find a ready market inside the country itself. This will incidentally minimize the import of citrus oils. There appears to be a bright future for the development of this industry in India.

In other countries, considerable amount of work has been done on the various methods of extraction of mandarin oils as affected by various factors like variety, maturity, and season[3, 17, 19], and on its physico-chemical composition and other aspects[2, 4-6, 9-14, 18, 20, 22, 24, 25, 30, 32] but in India, very little literature is available[1, 21, 26, 28]. Paul[27] conducted extensive studies on the effect of storage temperature, light, moisture and other factors on some citrus oils other than mandarin oils. Incidentally, no literature is available on the cold-pressed Nagpur mandarin oil. The results of analysis of Nagpur mandarin oil of 1954 season (supplied by the *Central Hindustan Orange & Cold Storage Co. Ltd.*, Nagpur) and briefly reported elsewhere[28] are presented in detail in this paper. The data are compared with the available data on other Indian and foreign mandarin oils.

Methods—The analysis of oil was conducted according to the methods of Guenther[15] and the results are presented in Table 2.

While surveying the literature on mandarin oils, the authors did not find any reference giving the quantitative data on the colour of these oils, the colour being usually expressed as light or deep orange. The colour of Nagpur orange oil was, therefore, measured in a Lumetron Photo-electric colorimeter at 6 different wave-lengths ranging from 650 μ to 420 μ. The observations were actually made as per cent light transmission, which were later converted to optical density and are presented in Table 3. For comparative easier appraisal, the colour was also measured in a Lovibond tintometer and the results are given in Table 4.

TABLE 1—IMPORTS OF CITRUS OILS IN INDIA

YEAR	LEMON OIL		BERGAMOT OIL		TOTAL IMPORTS	
	Qty. *gal.*	Value *Rs.*	Qty. *gal.*	Value *Rs.*	Qty. *gal.*	Value *Rs.*
1944-45	475	53,369	286	54,991	761	1,08,360
1945-46	1,887	2,17,065	726	1,89,263	2,613	4,06,328
1946-47	3,144	3,55,441	820	1,82,219	3,964	5,37,650
1947-48	2,897	3,17,026	2,481	2,63,020	5,378	5,80,046
1948-49	1,410	1,36,998	235	36,503	1,645	1,73,501
1949-50	2,183	1,70,561	582	75,921	2.765	2,46,482
1950-51	1,980	2,00,909	903	1,71,215	2,883	3,72,124
1951-52	1,428	3,05,087	580	1,41,732	2,008	4,46,819
1952-53	659	1,28,955	252	1,13,184	911	2,42,139

TABLE 2—SOME PHYSICO-CHEMICAL CHARACTERISTICS OF COLD-PRESSED MANDARIN PEEL OILS

	Indian mandarin peel oils				Foreign mandarin peel oils		
	Nagpur mandarin		Sylhet mandarin[26]	Coorg mandarin[26]	Italian tangerin[11]	Brazilian mandarin[15]	Florida tangerin[18]
	Sample I	Sample II					
Sp. gr.	0.8462 (25/25°C)	0.8472 (25/25°C)	0.8413 (18°C)	0.8553 (18°C)	0.854-0.859 (15°C)	0.854-0.858 (15°C)	0.845 (25°C)
$[\alpha]_D$	+100.7 (Alc. soln.)	+100.0 (1.25)(30°C)	+113.3 (18°C) (chloroform)	+114.3 (18°C)	+65°0′-+75°0′	+63°55′-+74°42′	+91.18 (25°C)
Refractive index	1.4745 20°C	1.4758 20°C	1.4730	1.4730	1.475-1.478	1.4745-1.4763	1.4734 (20°C)
Evaporation residue, %	2.99	3.60	2.0	2.57	2.4-3.5	3.4-4.7	4.53
Acid number	1.77	1.59	..	..	up to 2.7	..	
Aldehyde value (expressed as decylaldehyde) (hydroxylamine method), %	0.37	0.32	..	..	..	0.4-1.5	1.08
Ester number, %	4.64	4.12	..	..	5.0-11.0	1.5-3.5	0.34
Flavour	Pleasant (normal)	Pleasant (slightly inferior to I)	..	..	..	..	..
Solubility in 95% alc.	Clearly soluble ; soluble in 5-9 vols. 90% alc. with slight haziness		..	..	10 of 90 with turbidity ; 1.5 of 95 not always clearly	Incompletely in 90	
Colour	Deep yellow with reddish tinge	Deep yellow with reddish tinge	..	..	Bluish fluorescence when diluted with alcohol	Deep green	Deep reddish orange

TABLE 3—PHOTO-ELECTRIC COLORIMETRIC ANALYSIS OF COLD-PRESSED NAGPUR ORANGE OIL*

	Wave-lengths μ					
	650	580	550	530	490	420
Sample A	.097	.108	.337	.475	.620	1.30
Sample B	.119	.137	.393	.538	.710	1.40

* Measured in a Lumetron photo-electric colorimeter and expressed as optical density.

TABLE 4—COLOUR OF COLD-PRESSED NAGPUR ORANGE OIL*

	Lovibond units		
	Red	Yellow	Blue
Sample A	3.0	20.0	0.0
Sample B	3.0	20.1	0.0

* As observed in Lovibond tintometer

Table 2 reveals some variations in the quality of different mandarin oils. Variety of fruit, degree of maturity and storage of fruit prior to the extraction of oil are the main factors which affect significantly the chemical but not the physical properties of the expressed citrus oils[19]. The quality of citrus peel oils, as indicated by their physico-chemical characteristics, is also influenced by the yield of oil obtained in any commercial process regardless of the type of extraction equipment employed. Thus with an increase in the yield of an oil, the values of specific gravity, evaporation residue and refractive index also increase, while the optical rotation decreases, since, by increasing the yield by drastic extraction methods, more high-boiling, high-molecular weight constituents are evidently extracted, which accordingly cause a reduction in the percentage of d-limonene (the most optically active constituent of citrus oils) resulting in the lowest optical rotation value[19]. Normally, the d-limonene content of Indian mandarin oils varies from 90-94 per cent[25].

Flavour and aldehyde value—The characteristic odour of mandarin oil is due to the presence of methyl anthranilate and methyl N-methyl anthranilate[7]. The methyl anthranilate content of hand-pressed Assam mandarin oil, cold-pressed Coorg mandarin oil, distilled Nagpur mandarin oil is 0.2, 0.12, 0.8 per cent respectively[26]. In trade, both these compounds are used wherever synthetic orange flavour is desired[7]. From the flavour standpoint, aldehydes also constitute one of the dominant factors in orange and other citrus oils. The two samples of Nagpur orange oil analysed seem to be rather poor in aldehyde content (Table 2), which may perhaps be due to a large amount of aqueous phase coming in contact with the oil during processing, as it has been reported that aldehyde content decreases as the amount of aqueous phase (coming in contact with the oil during processing) is increased[19]. Thus, to produce an oil of high aldehyde content, the amount of aqueous phase should be reduced to the minimum under the operating conditions.

Evaporation residue of Brazilian and Florida oils is higher than those of Indian and Italian oils. This may be due to the fact that Italian oils were made by hand pressing while the Brazilian ones by machine pressing[13]. The slightly high figure for the Nagpur orange oil may be explained likewise ; it being machine pressed. This fact also reflects on the colour ; the Italian oil being lighter.

Ester value—It will be seen that the ester content of Nagpur orange oil is lower than that of Italian oil, but is higher than that of Florida oil (Table 2).

Colour—Tables 3 and 4 clearly depict quantitatively the nature and concentration of the colour of the two samples of Nagpur orange oil. Though the colour analysis by Lovibond tintometer (Table 4) does not reveal any significant difference, between the colours of two samples, the photo-electric colorimetric analysis (Table 3) clearly shows that sample B is comparatively darker in colour.

In both the cases, the optical density (colour absorption) was minimum at 650μ and maximum at 420μ, showing predominance of yellow and red colour in the oil. It will be of interest, however, to study the effect of various factors like storage temperature, light and moisture, on the stability of this oil and its colour, as hitherto no work seems to have been reported on this aspect. The use of suitable processing methods, proper packaging and storage under suitable conditions will enhance the shelf-life of these oils.

U.S. specification for tangerine oil—The Food Distribution Administration, U.S. Department of Agriculture, Washington. D.C. (1943) suggest the following specifications for tangerin oils[12]: sp. gr.$^{25°}$ 0.846-0.852 ; $[\alpha]_D^{25°}$, $+90°0' \div 95°0'$; $n^{20°}$, 1.4755-1.4769 ; colour, bright to deep orange. These limits evidently differ considerably from those of Italian and Brazilian mandarin oils, while all the Indian mandarin oils reported in Table 2 approach almost close to these specifications.

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Chemical Examination of Some Indian Petitgrain Oils

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The physico-chemical properties of oils extracted from *Citrus karna* and *Citrus limettioides* have been examined with regard to their suitability for perfumery purposes.

Petitgrain oils are obtained from the leaves and twigs of various citrus plants. They, along with the bergamot, orange, lime, lemon and mandarin oils extracted from the peels and neroli oils recovered from the flowers of sweet and bitter oranges, are extensively, used in the flavour and perfumery trades. Originally the name 'petitgrain oils' denoted only the oils obtained from twigs and leaves of bitter oranges, but now, similar oils from other citrus plants are also commonly designated as petitgrain oils.

Petitgrain oils are yellowish in colour and have an aromatic bitter taste. They are blended with ylang ylang, ionone and coumarin for use as perfume in skin creams ; they are also used for perfuming cheap soaps.

The industrial extraction of petitgrain oils was first started by Bolanza[1], a French botanist, who later introduced it in Paraguay, where bitter oranges grow wild.

The yield of the oil is reported to depend, besides other factors, on the conditions of planting, leaf material, season and weather. It varies from 0.25 to 0.5 per cent on the weight of green leaves. Leaves from young and healthy trees yield more oil than those from old ones. The leaves from the trees, which have been trimmed in the preceding year, give a better yield. There is a direct relationship between the yield and quality of the oil ; a high yield indicating a high quality[2]. The oil content in the leaves diminishes as the season advances. The freshly-cut leaves yield more oil (0.280 per cent) than those which are stored for three days (0.225 per cent). It was further observed that chopped leaves yield less oil (0.260 per cent) than whole leaves (0.285 per cent). It is believed that this abnormality is due to the bumping of the leaves in the still which results in the hydrolysis of esters.

It has been observed that: (1) Yield of oil from leaves of sweet oranges is lower than that from leaves of the bitter variety ; (2) leaves from trees grown in plains give a lower yield and inferior quality of oil than those grown in hills ; (3) the highest yield and quality of oil are obtained from January to June

TABLE 1—CONSTITUENTS AND PHYSICO-CHEMICAL CONSTANTS OF SOME PETITGRAIN OILS

No.	PETITGRAIN OILS	YIELD %	CONSTITUENTS	SP. GR.	$[\alpha]_D$	n	ACID VAL.	ESTER VAL.	ALDEHYDES %
1.	*Citrus aurantifolia*[2] Limes (West Indies)	. .	. .	0.877-0.878	−37° to −38°	. .	3-5	22-25	43
2.	***Citrus aurantium***[4] Bitter orange (France)	Up to 0.4	Linalool, nerol, geraniol, terpinol, and their acetic esters ; methyl anthranilate, camphene, pinene, limonene and furfural	0.888-0.897	−1° to −8°	1.460 to 1.470	. .	50-70	. .
3.	*Citrus decumana*[5]	. .	Linalyl acetate 44.18% Linalool 42.34% α-β-pinene 6.60% d-α-pinene 2.60%	. .	. .	. .	. .	. .	. .
4	*Citrus paradisi*[3] Grape fruit (Philippines)	1.7	Dipentene & linalool 15% Citral 1%	0.870†	22.9°†	1.4644†	. .	10	. .
5.	Sweet orange[6] (Brazil)	0.1	. .	0.8962*	11.2°*	1.4511*	. .	9.4	15.8

* At 25°C.
† At 30°C.

and then fall off gradually ; the poorest yield is obtained from August to October ; (4) better yield and quality of oil are obtained from airy and sunny plantings than from those in dense forests and jungles ; and (5) distillation of fresh leaves should be carried out as rapidly as possible to get superior quality of oil and high yield.

The physico-chemical constants of some of the petitgrain oils are given in Table 1.

Very little work seems to have been done in India on petitgrain oils, although India stands sixth[7] in the world in the production of citrus fruits. Nigam and Dutt[5] have examined the oil from the leaves of *Citrus decumana* and Rao *et al.*[8] and Nayak and Guha[9] examined the petitgrain oils from *Citrus medica* Linn, var. *acida* and wild limes (*Atlantia monophylla* Correa) respectively. The analytical results are given in Table 2.

Terpeneless petitgrain oils are very important perfumery raw materials. They are prepared by fractional distillation in such a manner that the constituents of low perfume value are removed leaving the oil stronger in characteristic perfume. Their increased solubility and accentuated characteristic aroma give an advantage over natural oils. The characteristics of the terpeneless petitgrain oil prepared by *Fritzsche Bros. Inc.*, New York during the past twenty years are given in Table 3.

As the citrus oils do not maintain the same quality on storage, it has been recommended that they be kept in tin-lined, fully filled drums at 65° F. Guenther[2] has suggested the simple method of placing a small quantity of dry ice (solid carbon dioxide) in the container. As the dry ice rapidly evaporates and drives out the air, it ensures the presence of neutral gas before the drum is tightly sealed.

India consumes large quantities of citrus oils as perfumes for soaps and cosmetics and as flavouring agents for aerated waters, beverages and confectionery. The value of citrus oils imported into the country is about Rs. 3.5-4.1 lakhs[10] per annum.

As Uttar Pradesh abounds in citrus plants, it was proposed to extract the oils from leaves of the two varieties, *Citrus karna* and *Citrus limettioides*, grown in our gardens and examine them with regard to their suitability for perfumery purposes.

PETITGRAIN OIL FORM *Citrus karna*

Citrus karna, known as *khatta*, *idnembu*, or *karna khatta* in Hindi, has no English name although it is frequently referred to as citron which it in no way resembles. The tree is very vigorous and is commonly and successfully used as stock. In some ways, the fruit resembles sour orange. Its flowers are tinged with red like those of lemon, but the leaves are distinct from those of either species, having neither the large wings of sour orange nor the distinct scent of lemon. The fruit is large and round but generally mammillate and sometimes very rough. Both the rind and pulp are orange in colour. The juice is plentiful and very sour.

TABLE 2—YIELDS AND PHYSICO-CHEMICAL CONSTANTS OF SOME INDIAN PETITGRAIN OILS

No.	Petitgrain oil	Yield %	Sp. gr.	n	$[\alpha]_D$	Acid val.	Ester val.	Sap. val. after acetylation	Solubility in 90% alcohol vol.
1.	*Citrus decumana*	. .	0.8849*	1.4635*	+4.5°	10.6	126.4	264.5	. .
2.	*Citrus medica* Linn. var. *acida*	. .	0.8744†	1.478†	. .	5.5	4.4	60	0.5
3.	*Atlantia monophylla*	0.4 to 0.6	0.8675†	1.465‡	−34.18°	6.85	92.86	139.9	1

* At 20°C.
† At 30°C.
‡ At 31.5°C.

TABLE 3—CHARACTERISTICS OF TERPENELESS PETITGRAIN OILS PREPARED BY FRITZSCHE BROS., NEW YORK

Sp. gr.$^{15°}$	0.889 to 0.898
$[\alpha]_D$	−0.55° to −2°58′
$n^{20°}$	1.4586 to 1.4619
Ester content (calculated as linalyl acetate), %	41.6 to 60.4
Solubility in 70% alcohol, vol.	2-3

TABLE 4—PHYSICO-CHEMICAL PROPERTIES OF PETITGRAIN OIL OBTAINED FROM CITRUS KARNA

	Sample 1	Sample 2
Sp. gr.$^{30°}$	0.8603	0.8595
$n^{30°}$	1.4690	1.4685
$[\alpha]_D$	+48°24′	+43°12′
Acid val.	5.71	1.47
Ester val.	35.91	47.3
Ester val. after acetylation	125.67	142.3
Esters, as linalyl acetate, %	12.56	16.4
Free alc., as $C_{10}H_{18}O$, %	26.46	28.13
Tertiary terpene alc., by formylation in cold, %	19.49	20.61
Aldehydes, as citral (sod. bisulphite method), %	19.5	18.1
Solubility in 90% alcohol, vol.	0.5	0.5

TABLE 5—TEMPERATURE RANGE AND YIELD OF PETITGRAIN OIL FROM CITRUS KARNA

FRACTION NO.	BOILING POINT °C.	TEMPERATURE OF OIL BATH °C.	WEIGHT OF THE FRACTION g.	YIELD %
1.	65- 68	96-100	35.670	27.872
2.	68- 70	100-105	13.220	10.330
3.	70- 73	105-110	17.510	13.680
4.	73- 76	110-120	8.245	6.442
5.	76- 85	120-130	8.313	6.495
6.	85- 90	130-135	3.159	2.468
7.	90-104	135-140	15.310	11.960
8.	104-107	140-145	9.506	7.427
9.	Residue	up to 180	15.255	11.910
	Loss		1.812	1.416
			128.00	100.00

A weighed quantity of fresh green leaves and twigs was distilled in a steam-heated still. The oil was separated from water and dried over anhydrous sodium sulphate. It was pale yellow in colour and had an odour like lemon oils (yield, 0.256 per cent in July and 0.286 per cent in December). The physico-chemical properties of the two samples of oils prepared are given in Table 4.

Sample 1 (128.0 g.) was fractionated at 10-11 mm. The results of fractionation are recorded in Table 5.

The fractions were individually analysed. The analytical data are given in Table 6.

Fractions 1 and 2, which appeared to contain mainly terpenes, were mixed (38.5 g.) in the same proportions as obtained in the fractionation of the oil and refractionated at 5-6 mm. over metallic sodium. The results of fractionation are recorded in Table 7.

Fractions A, B and C did not show the presence of free acids, esters and aldehydes. They had sp. gr.$^{30°}$, 0.8405, 0.8435 and 0.8467 ; and $[\alpha]_D$, +83°15′, +85°15′, and +86°0′ respectively. These were identified mainly as *d*-limonene and dipentene. The presence of dipentene was confirmed by preparing its dihydrochloride, nitrosochloride and tetrabromide derivatives.

As the total weight of the fractions A, B and C was 26.78 g., the terpenes content of the combined fractions 1 and 2 was 69.55 per cent (26.57 per cent in the oil).

The high optical rotations and low specific gravities of fractions 3, 4 and 5 indicated that they also contained terpenes. They were combined (10.5 g.) in the same proportion as obtained from the original oil and refractionated over metallic sodium at 5-6 mm.

The distillate collected up to 61°C. weighed 1.56 g. and did not show the presence of free acids, esters and aldehydes. It showed $[\alpha]_D$, +85°24′.

TABLE 6—PHYSICO-CHEMICAL CONSTANTS OF PETITGRAIN OIL FRACTIONS FROM CITRUS KARNA

FRACTION NO.	SP. GR.	n^{30}	$[\alpha]_D$	ACID VAL.	ESTER VAL.		ESTER CONTENT AS LINALYL ACETATE %	ALCOHOLS AS $C_{10}H_{18}O$	ALDEHYDES AS CITRAL %
					Initial	After acetylation			
1	0.8467	1.4675	+74°57′	4.75	15.12	46.03	5.28	8.69	—
2	0.8475	1.4695	+73°0′	2.64	21.80	58.87	7.62	10.47	8.56
3	0.8556	1.4680	+69°36′	3.75	24.61	178.00	8.606	47.63	9.84
4*	0.8584	1.4680	+64°20′	10.30	54.58	214.50	19.08	49.65	13.32
5*	0.8623	1.4685	+48°36′	15.10	56.14		19.63		17.34
6	0.8674	1.4690	+28°20′	6.93	17.72	—	6.197	—	36.40
7	0.8717	1.4690	+10°48′	5.00	47.83	175.20	16.73	38.17	40.12
8	0.8814	1.4695	+3°24′	1.40	60.99	152.30	21.32	26.95	46.47
9	Residue	1.4920	—	9.03	68.79	—	24.06	—	51.30

* Fractions 4 and 5 were mixed and then acetylated.

TABLE 7—REFRACTIONATION OF MIXTURE OF FRACTIONS 1 AND 2 OF PETITGRAIN OIL FROM CITRUS KARNA

Fraction No.	Boiling point °C.	Temperature of oil bath °C.	Weight g.	Yield %
A	46-50	60-70	6.704	17.42
B	50-54	70-80	9.098	23.63
C	54-57	Up to 100	10.978	28.52
Loss due to formation of sodium compounds or otherwise			11.720	30.43
		Total	38.50	100.00

TABLE 8—APPROXIMATE COMPOSITION OF PETITGRAIN OIL FROM CITRUS KARNA

Constituents	%
Terpenes (d- and dl-limonene)	30.56
Tertiary alcohols (mainly d-linalool)	19.49
Primary alcohols (mainly geraniol)	6.97
Citral	19.50
Esters, calculated as linalyl acetate	12.56

It was taken as terpene, 14.85 per cent in the combined fraction (3.99 per cent in the oil). This makes the total quantity of terpenes present in the oil, 30.56 per cent.

Fraction 3 was found to contain 47.63 per cent free alcohols as geraniol besides 9.837 per cent aldehydes as citral. After the removal of aldehydes with potassium meta-bisulphite, the residue showed the presence of linalool, which was identified through its phenyl urethane (m.p. 65°C.).

Fractions 4 and 5 contained 13.32 and 17.34 per cent aldehydes (as citral) respectively. The aldehydes were identified to be citral through their semicarbazones.

Fractions 4 and 5 were combined and examined for their ester value after acetylation. Free alcohol content was found to be 49.65 per cent ; the alcohol was identified as linalool.

Fraction 6 being small, could not be investigated for its ester value after acetylation. However, the presence of linalool was qualitatively detected in it.

Fractions 7 and 8 contained citral 40.12 and 46.47 per cent respectively. Their ester values after acetylation were determined separately and the alcohol contents were found to be 36.12 and 26.95 per cent respectively. These fractions were combined and after the removal of aldehyde were mixed thoroughly with 9 g. of finely pulverized anhydrous calcium chloride. The resulting complex was washed with petroleum ether and decomposed with water ; the liberated oil was steam distilled. The distillate was confirmed to be geraniol, through its diphenyl urethane (m.p. 82.5°C.).

Fraction 9 (residue) contained 51.3 per cent citral along with 24.06 per cent of esters. It has an acid value of 9.03 and did not contain citroptene.

The ester contents of all the fractions were calculated as linalyl acetate. Methyl anthranilate could not be detected in any of the fractions or in the original oil.

Composition

The approximate composition of the oil as determined in these investigations is given in Table 8.

TABLE 9—PHYSICO-CHEMICAL PROPERTIES OF PETITGRAIN OIL OBTAINED FROM CITRUS LIMETTIOIDES

	SAMPLE 1	SAMPLE 2
Sp. gr.$^{30°}$	0.8716	0.8704
$n^{30°}$	1.4727	1.4735
$[\alpha]_D$	+38°42′	+40°30′
Acid val.	8.96	6.9
Ester val.	55.42	53.98
Esters, as linalyl acetate, %	19.38	18.9
Ester val. after acetylation	143.3	139.9
Free alc. as geraniol, %	25.86	25.28
Tertiary terpene alc. by Glichitch's method for formylation, %	16.42	—
Aldehydes, as citral, %	12.80	13.7
Solubility in 90% alc., vol.	0.5	0.5

TABLE 10—YIELDS OF VARIOUS FRACTIONS OF PETITGRAIN OIL FROM CITRUS LIMETTIOIDES

FRACTION NO.	BOILING POINT °C.	WEIGHT g.	YIELD %
1	54- 60	13.792	13.061
2	60- 64	18.428	17.451
3	64- 73	11.552	10.940
4	73- 84	6.924	6.557
5	84- 95	4.876	4.617
6	95-105	10.924	10.345
7	105-112	7.328	6.940
8	112-113	13.444	12.729
9	Residue	16.444	15.572
	Loss	1.888	1.788
		105.600	100.000

TABLE 11—PHYSICO-CHEMICAL CONSTANTS OF PETITGRAIN OIL FRACTIONS FROM CITRUS LIMETTIOIDES

FRACTION NO.	SP. GR.$^{30°}$	$n^{30°}$	$[\alpha]_D$	ACID VAL.	ESTER VAL.		ESTER CONTENT AS LINALYL ACETATE %	ALCOHOLS AS $C_{10}H_{18}O$ %	ALDEHYDES AS CITRAL %
					Initial	After acetylation			
1	0.8464	1.4704	+75°36′	3.4	20.98	—	7.336	—	—
2	0.8480	1.4709	+74°24′	4.6	23.09	81.96	8.170	16.92	6.640
3	0.8504	1.4724	+67°12′	5.98	24.36	93.45	8.521	20.05	7.216
4	0.8752	1.4729	+50°24′	11.4	54.92	186.74	19.220	40.09	10.160
5	—	1.4719	+46°12′	11.5	65.56	204.06	22.930	42.48	20.420
6	0.8772	1.4714	+4°48′	5.75	67.23	195.83	23.520	39.11	24.750
7	0.8785	1.4724	+1°12′	9.2	100.90	177.54	35.290	22.66	27.670
8	0.8796	1.4734	nil	10.77	72.36	203.4	25.310	40.05	21.200
Residue	0.8802	1.5302	—	21.1	92.70	182.02	32.450	26.31	13.820

Petitgrain Oil from *Citrus limettioides*

Citrus limettioides is known as *mitha lemu* or *mitha nembu* in Hindi, Bengali and Gujarati. It is probably of hybrid origin and is commonly grown in Central and Northern India. The fruit ripens late in the rainy season when other citrus fruits are not available.

Fresh green leaves of *Citrus limettioides* (196.3 lb.) distilled in a steam-heated still during May gave 160.5 g. (0.18 per cent) of a pale yellow oil with lemon-like odour. Its physico-chemical properties are given in Table 9.

The oil (105.6 g.) was fractionated at 10-12 mm. The yields of various fractions are given in Table 10.

The physico-chemical constants of the fractions are tabulated in Table 11.

Fractions 1, 2 and 3 with their high optical rotation and low specific gravity appear to contain terpenes. They were combined (20.732 g.) in the same proportions as obtained from the oil and refractionated over metallic sodium at 10-12 mm. The fractions collected are given in Table 12.

Fractions A and B did not contain free acids, esters or aldehydes. They showed $[\alpha]_D$, +84°30′ and +87.0′ respectively indicating the presence of terpenes. *d*-Limonene and *dl*-limonene (dipentene) were identified through their dihydrochloride, tetrabromide and nitrosochloride derivatives. The quantity of terpenes in fractions 1, 2 and 3 was 63.8 per cent (26.46 per cent in the oil).

Fraction 4 was so small that no oil was left for the determination of terpenes. It contained: aldehydes, 10.16 per cent and alcohol, 40.09 per cent. The alcohol was identified as linalool.

TABLE 12—REFRACTIONATION OF PETITGRAIN OIL FRACTIONS 1, 2 & 3

Fraction no.	Boiling point °C.	Weight g.
A	51-64	5.875
B	64-68	7.360
Loss		7.497
	Total	20.732

TABLE 13—APPROXIMATE COMPOSITION OF PETITGRAIN OIL FROM CITRUS LIMETTIOIDES

Constituents	%
Terpenes (mainly d-limonene and dl-limonene)	26.5
Linalool	16.42
Geraniol and nerol	9.44
Esters, as linalyl acetate	19.38
Citral	12.89
Citroptene	Traces

Fraction 5 contained 20.42 per cent aldehydes as citral. As the fraction was small, the alcohol could not be identified. The presence of linalool, however, was detected by the Liebermann Burchand reaction.

Fraction 6 contained 24.75 per cent citral. After the removal of citral, the linalool content of the fraction was estimated and found to be 39.11 per cent.

Fractions 7 and 8 contained citral 27.67 and 21.2 ; and alcohols, 22.66 and 40.05 per cent respectively. The fractions were mixed together after removal of the aldehyde ; the mixture (10 cc.) along with 8 g. of phthalic anhydride and 10 cc. of benzene was refluxed for 2 hours on a water bath. The excess of phthalic anhydride was separated with petroleum ether. The solvent was removed and the residue shaken with sodium carbonate solution. The aqueous solution was acidified with 20 per cent sulphuric acid and the acid phthalate extracted with ether. It was saponified by refluxing with alcoholic potash ; the aromatic alcohol was liberated by adding water, and extracted with ether. It had a pleasing rose like odour. The presence of geraniol was confirmed through its diphenyl urethane.

It was observed that some of the oil did not react with calcium chloride. This along with calcium-geraniol compound was extracted with ether. The residual liquid, which after the removal of ether was very small in quantity, boiled at 224°C. and showed n^{30°, 1.4703 indicating the presence of nerol.

The residue left after fractionation contained 13.82 per cent aldehydes and 22.6 per cent alcohols. Traces of citroptene were also detected.

The approximate composition of the oil as determined through these studies is given in Table 13.

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An Evaluation of Some Indigenous Perfume Bearing Materials

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The essential oils from nine different raw materials, namely, the buds, chips, featherings and twigs of cinnamon, the rhizomes of *Gorabach* and *Kapur kachri*, the roots of *Jatamansi* and *Kuchu gundubi* and the berries of *Sugandha kokala*, have been obtained by the process of steam distillation. In addition to determining the physical, chemical and odoriferous characteristics of the oils, the economics of the distillation process have been included.

***Gorabach*, *Kapur kachri* and *Sugandha kokala* are rich in oil content, and these oils possess good odoriferous characteristics. *Kuchu gundubi* is distinctive in as much as it is the only material exhibiting flowery notes whereas the other oils have spicy or heavy non-distinctive notes.**

For centuries past, India has been recognized as a land rich in aromatic materials. Even in the Vedas, which are over 3,000 years old, cosmetics and perfumes used in honour of divinity are referred to with admirable knowledge and detail. Although India is rich in aromatic materials, it has remained primitive in the techniques of their exploitation and application. With the passage of time, there has undoubtedly been a change in accent. Thus, in the past, the materials which were valued greatly were the flowers of mango, champak, hyacinth, rose, violet, lily and narcissus as well as the resin of benzoin and musk and roots of vetiver whereas the materials which are currently exploited on a commercial scale are sandalwood, vetiver, lemongrass, palmarosa, gingergrass, cinnamon, terebinth, etc.

In addition to the essential oils enumerated above, there are many perfume-bearing materials available in India, whose essential oils are not being distilled on a commercial scale. Until about a decade ago, there was no authentic information relating to these minor perfume-bearing materials but, during the last few years, the various State Forest Departments and institutions, notably the Forest Research Institute, Dehra Dun, have started investigations relating to the survey of these materials. A few trial distillations have been conducted by the authors in order to determine whether the essential oils derived from the buds, chips, featherings and twigs of *Cinnamomum zeylanicum* Breyn. (*Dalchini*), rhizomes of *Acorus calamus* Linn. (*Gorabach*) and *Hedychium spicatum* Ham. (*Kapur kachri*), roots of *Nardostachys jatamansi* D.C. (*Jatamansi*) and *Homalomena rubescens* Kunth. (*Kuchu gundubi*) and the berries of *Luvunga scandens* Ham. (*Sugandha kokala*) could be utilized advantageously in perfume compositions.

EXPERIMENTAL

Raw material—Cinnamon trees are grown in Mysore, South Kanara and South Malabar. Cinnamon oil is distilled principally from the bark of the tree ; the buds, chips, featherings and twigs, are disposed off by burning. Samples of these four materials were obtained from Brown's Cinnamon Estate, Anjarkandy, South Malabar. *Gorabach, Jatamansi, Kapur kachri* and *Sugandha kokala* are reported to be common in the Kumaon Range of the Himalayan forests. The rhizomes of *Gorabach* and *Kapur kachri* as well as *Jatamansi* roots and *Sugandha kokala* berries are collected from the Himalayan forests and brought to Kathgodam from where they are despatched to Bombay, Calcutta and Kanauj for use in Ayurvedic preparations and perfumery. Samples of these materials were obtained from the Range Forest Officer, Pundu Range, Thornli, Garhwal. *Kuchu gundubi* is reported to be fairly common in Sikkim and in various parts of Assam. The roots were obtained from Messrs *Ashok Brothers Ltd.*, Silchar, Assam. Some particulars of these raw materials are given below:

Cinnamon buds—These are greyish black in colour and resemble Zanzibar clove buds to a certain extent.

Cinnamon chips—These consist of small pieces, $\frac{1}{2}$ to 1 in. long, $\frac{1}{4}$ to $\frac{1}{2}$ in. wide, and 0.025 to 0.05 in. thick. The chips are coloured grey on one side and brownish yellow on the other, indicating that they form the outermost core of the cinnamon tree.

Cinnamon featherings—These are curled pieces, $2\frac{1}{4}$ to 4 in. long and 0.125 to 0.775 in. in diam., and are brown in colour.

Cinnamon twigs—This material consists of yellowish brown sticks, $\frac{1}{2}$ to 4 in. long, and approximately 0.05 in. thick. They are accompanied by broken buds and leaves.

Gorabach rhizomes—This material consists of brown coloured rod-shaped rhizomes, about 4 in. long, which are covered with numerous scale leaves.

Kapur kachri rhizomes—These are disc-like rhizomes, $\frac{3}{4}$ to $1\frac{1}{4}$ in. in diam. and about 1/8 in. in thickness. The colour of the material is darker and more greyish towards the edges ; the middle portion being near-white. The rhizomes are porous and the surface is powdery.

Jatamansi roots—These are about 1.5 to 2 in. long rod-shaped hollow roots with a profuse hairy growth on the surface.

Kuchu gundubi roots—These are small brown coloured rod-shaped roots, $\frac{1}{2}$ to 1 in. in length and $\frac{1}{4}$ in. in diam.

Sugandha kokala berries—These are brown and black berries, $\frac{1}{4}$ to $\frac{1}{2}$ in. in diam., with a wrinkled outer surface. The outer hard coat encloses a relatively soft seed.

Distillation—The trial distillations were conducted using an experimental steam distillation unit of 10 gal. capacity. Prior to charging in the still, the

TABLE 1—ESTIMATES OF COST OF PRODUCTION OF SOME INDIGENOUS ESSENTIAL OILS

	CINNAMON								
ITEMS	Buds	Chips	Featherings	Twigs	*Gorabach*	*Jatamansi*	***Kapur** kachri*	*Kuchu gundubi*	*Sugandha kokala*
Estimated quantity of raw material available, lb.	10,000 to 12,000	20,000 to 30,000	12,000 to 20,000	2,000	40,000	20,000	48,000	20,000	15,000
Moisture content, %	12.5	12.4	13.1	13.3	12.5	6.7	12.2	12.8	7.7
Yield of essential oil, %	0.40	0.17	0.55	0.07	4.60	0.58	1.00	0.15	4.18
Quantity of essential oil which could be distilled, lb.	40-48	33-50	65-100	1.5	1,850	160	475	30	650
Cost of raw material per ton, Rs.	2,300	840	2,800	1,120	800	2,300	700	1,120	1,500
Approximate cost of processing raw material per ton, Rs.	115	60	120	55	760	730	450	250	200
Approximate cost of essential oil per lb., Rs.	270	240	240	800	16	235	50	410	20

raw materials were reduced to the desired size in a disintegrator. During the distillation runs, various particulars, such as the rate of distillation, the actual content of volatile oil in the distillate, the consumption of steam and water, etc., were noted with a view to arriving at the cost of processing the raw materials. Some of the more important data are presented in Table 1.

RESULTS AND DISCUSSION

The essential oils obtained by steam distillation of various raw materials have been examined in respect of their physical and chemical characteristics, and the data obtained are summarized in Table 2.

Cinnamon bud oil—This is a greenish yellow liquid with a relatively sweet spicy odour. Its cinnamic aldehyde content is low, (about 3 per cent) and it cannot be used to replace cinnamon bark oil. The intrinsic odour of cinnamon bud oil is quite interesting, but since the total quantity of the oil available is less than 50 lb. per annum and since its cost is rather prohibitive, it appears as though it would not be an economic proposition to distil it commercially.

Cinnamon chip oil and feathering oil—These are yellow to brown liquids with spicy odour, reminiscent of cinnamon bark oil. The cinnamic aldehyde contents of the oils derived from the chips and featherings are about 52 per cent of cinnamic aldehyde. The spicy odour of this oil is slightly stronger than that of the bark oil, both these oils are good essences and could be utilized as substitutes for the bark oil. If the oils derived from the chips and featherings are mixed, about 100 to 150 lb. of essential oil would be available per annum but, as in the case of the bud oil, the cost of these materials is prohibitive from the point of view of their commercial utilization.

Cinnamon twig oil—This is a greenish yellow liquid containing about 8 per cent of cinnamic aldehyde. The spicy odour of this oil is slightly stronger than that of the bud oil, but in view of the extremely small quantity available, it would not be an economic proposition to utilize this raw material.

Gorabach oil—This is a yellow liquid with a sweet but slightly nauseating odour, its fragrance being vaguely reminiscent of patchouli. It appears as though this oil, upon fractionation, will yield a material of good perfumery value. If all the raw material which is available currently is used for distillation, approximately 1850 lb. of the essential oil can be distilled per annum. The price of the oil is reasonably cheap, and in view of the distinct possibilities shown by its odoriferous characteristics, it appears as though the utilization of this raw material would be quite profitable.

Jatamansi oil—This is a dark brown liquid with a natural musky note of high intensity. Although its odour may be considered by some to be disagreeable, proper fractionation of the essential oil should yield a material of considerable perfumery value. On the other hand, the total quantity of the essential oil available is relatively small, and its cost is very prohibitive.

Kapur kachri oil—This is a brownish yellow liquid with a camphoraceous odour and a gummy balsamic note. To a certain extent, its odour is

TABLE 2—PHYSICAL AND CHEMICAL PROPERTIES OF SOME INDIGENOUS ESSENTIAL OILS

ESSENTIAL OIL		SP.GR.$^{15°}$	$n^{20°}$	OPTICAL ROTATION $[\alpha]_D$	SOLUBILITY AT 20°C. IN VOL. ALC., %	ACID VAL.	ESTER VAL.	
							Initial	After acetylation
Cinnamon	buds	0.9398	1.5030	+12°27′	9 of 90	6.5	7.1	49.1
	chips*	0.9996	1.5623	−0°45′	1.5 of 80	9.9	. .	. .
	featherings**	1.0139	1.5798	−1°9′	1 of 80	3.1	. .	. .
	twigs	0.9333	1.4965	−0°54′	1 of 80	5.6	19.5	. .
Gorabach		1.0889	1.5623	0°	1.5 of 70	1.5	2.9	29.6
Jatamansi		0.9731	1.5038	Not determined.	Not completely soluble even in 10 of 90	8.6	51.5	77.9
Kapur kachri		0.9441	1.4898	−1°24′	1 of 80	2.2	9.2	104.6
Kuchu gundubi		0.9002	1.4710	−9°24′	2 of 70	12.8	5.3	132.5
Sugandha kokala		0.9289	1.4918	−16°18′	1.5 of 90	9.2	75.6	112.3

* Cinnamic aldehyde content, 52.5 per cent.

** Cinnamic aldehyde content, 65.5 per cent.

reminiscent of zedoary oil which, in turn, is suggestive of tuberose. This oil is relatively plentifully available and its price is moderate ; therefore, it has distinct possibilities.

Kuchu gundubi oil—This is a yellowish brown liquid with a predominant smell of terpineol and a back note of linalool. It appears as though fractionation of this oil can yield a material which would be rich in linalool content and thus has a pleasant flowery odour. The total quantity of the essential oil available is relatively small and its cost is prohibitive.

Sugandha kokala oil—This is a yellow liquid with a sweet camphoraceous odour, reminiscent of spike lavender. As this oil is available in moderate quantities at a reasonably cheap price, it has distinct possibilities.

Investigation of Rosha Oils : Part II—Isolation of Geraniol from Rosha Oils

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Study of the isolation of geraniol from rosha oil through the formation of calcium chloride adduct shows that lower temperature (2°C.), longer time of contact (68 hr.) and finer particle size of calcium chloride (–50 mesh) increase the yield of geraniol in the Jacobsen method. In the method of Jones and Wood the time of contact is reduced to 2 hr. but the proportion of calcium chloride to oil is greatly increased. Adduct formation does not take place in solvents like ethyl ether, benzene, carbon tetrachloride and acetone. There is a rapid fall in the yield of geraniol with the decrease of the geraniol content of the oil. The addition product is apparently not formed when the geraniol content is *c*. 40 per cent.

Geraniol from palmarosa oil fetches a higher price than geraniol concentrates obtained from other oils like the Java citronella oil because of its sweet odour. Sofia oil (ginger grass oil) obtained from the same species of grass as palmarosa oil, is not in great demand as a perfume because of its low geraniol content (30-40 per cent) and penetrating odour caused by a high proportion of terpenes.

The common method of obtaining geraniol concentrates is the distillation of the raw oils under reduced pressure. Geraniol can also be isolated through its addition compounds formed with calcium chloride, calcium nitrate, magnesium chloride and magnesium nitrate. The calcium chloride adduct ($CaCl_2.2C_{10}H_{18}O$) is insoluble in petroleum ether and is easily decomposed by water liberating geraniol. Jacobsen[1] was the first to propose such a method for the isolation of geraniol in the year 1871. In this method which has been recently studied by Lal and Mathur[2], powdered anhydrous calcium chloride is intimately mixed with the oil and the paste so formed is kept at about 0°C. for 24 to 48 hr. The cold paste is then extracted with petroleum ether, filtered and washed several times with the solvent. The insoluble addition product is then decomposed with ice cold water to liberate the geraniol.

An important modification of the Jacobsen's method, in which the oil (Java citronella) was added to a finely powdered anhydrous calcium chloride suspended in hexane at room temperature, was suggested by Jones and Wood[3]. According to these workers, the adduct formation was complete in about 2 hr. Further separation of geraniol was carried out in the same way as in Jacobsen's method.

In a critical study of these two methods, the influence of several factors on the recovery of geraniol must be ascertained in order to arrive at optimum conditions. In the present work the following aspects have been studied.

(1) The Jacobsen method, which may be called the 'direct method', has been studied with respect to the effect of (*a*) time of contact (*b*) temperature of reaction and (*c*) fineness of calcium chloride. The free geraniol content of the oil, moisture content of the calcium chloride and the proportion of calcium chloride to oil were kept constant.

(2) With respect to the modified method of Jones and Wood[3] which may be called the 'solvent method', the effect of (*a*) time of stirring (*b*) temperature of reaction and (*c*) the fineness of calcium chloride has been studied. In both the cases, motia oil of a geraniol content of 79.5 per cent has been used.

(3) The solvent method has also been applied to sofia oil and factors such as geraniol content of the original oil, proportion of calcium chloride to oil, and amount of solvent, were varied, keeping constant the time of contact and temperature of reaction.

EXPERIMENTAL

Rosha oil samples—Motia oil with free geraniol content of 79.5 per cent and a volatile fraction having a free geraniol content of 12 per cent obtained by fractional steam distillation of sofia oils were suitably mixed in order to obtain oil samples with the desired concentration of geraniol.

Solvent—'Solvent Oil' from Messrs *Burmah Shell Co. Ltd.* was redistilled and the fraction, boiling between 65 and 68°C., similar to technical hexane, was utilized.

Calcium chloride—The calcium chloride used was heated at about 500-550°C. for 2 hr. to ensure complete removal of moisture, cooled to 300°C. and stored in a desiccator over phosphorus pentoxide.

Estimation of geraniol—Total alcohols, calculated as free geraniol, were estimated by determining the hydroxyl value. A mixture of pyridine and acetic anhydride was used as the acetylating reagent. The procedure has already been given in Part I of this paper[4]. Tables 1-4 give the results of the investigation.

In all the above experiments (Tables 1-4) the purity of the geraniol fractions isolated ranged from 97 to 98 per cent, the remaining 2 to 3 per cent being residual petroleum ether. The retention of 2 to 3 per cent of petroleum ether in the geraniol concentrate was confirmed by distilling off the solvent from a mixture of rosha oil and petroleum ether, and by further heating the mixture for about 10 minutes at 70-80°C. under a pressure of about 40 cc. of mercury. The increase in the percentage of rosha oil was found to be of the order of 2.

DISCUSSION

'Direct method' of isolating geraniol—It is seen (Table 1) that lower temperature (about 2°C.) and prolonged time of contact (nearly 68 hr.) tend

TABLE 1—INFLUENCE OF DIFFERENT FACTORS ON YIELD OF GERANIOL (DIRECT METHOD)

(Geraniol content of oil, 79.5% ; ratio of calcium chloride to oil, 1 : 1)

No.	REACTION TEMPERATURE °C.	TIME OF CONTACT hr.	SIZE OF THE CHLORIDE mesh	YIELD OF GERANIOL %
1	30	48	−50	42
2	2	48	−50	51
1	2	24	−50	41
2	2	48	−50	50
3	2	68	−50	59
4	2	72	−50	60
1	30	48	−28, +30	35
2	30	48	−50	42

TABLE 2—INFLUENCE OF TIME OF CONTACT AND SIZE ON YIELD OF GERANIOL (SOLVENT METHOD)

(Geraniol content of oil, 79.5% ; ratio of calcium chloride to oil, 2.4 : 1 ; temperature of reaction, 30°C. ; ratio of solvent to oil, 4 : 1)

No.	DURATION OF STIRRING hr.	SIZE OF THE CHLORIDE mesh	YIELD OF GERANIOL %
1	1	−50	56.0
2	2	−50	62.0
3	3	−50	63.5
4	4	−50	62.0
1	1	−28, +30	43.5
2	1	−50	56

to increase the yield of geraniol. Under the conditions studied, a yield of 60 per cent corresponding to a geraniol recovery of the order of 75 per cent was obtained. The weight ratio of calcium chloride to oil is 1 : 1 which is 200 per cent in excess of the theoretical requirement for the formation of the adduct. Since the reaction would take place only on the surface of the calcium chloride particles, the size of the chloride, besides the amount used, is also a governing factor.

'Solvent method' of isolating geraniol—The maximum geraniol yield of 63 per cent is obtained with an optimum time of 2 hr. (Table 2). Although the yield is comparable with the 60 per cent yield obtained in the 'direct method', the proportion of calcium chloride required is much more as indicated by the set of results given in Table 3. It may appear from Table 3 that the amount

of solvent used does not affect the yield of geraniol to a considerable extent. It is, however, observed that when the amount of solvent is reduced, the contents of the flask become very thick as the reaction proceeds, and it is likely that this may offset the slight improvement in the yield on reducing the amount of solvent. The effect of the solvent can be better seen in experiments 7 and 8 in Table 4.

Effect of geraniol content of original oil on the yield of geraniol—It is noticed from Table 4, Experiment 1, that if the original oil contains about 40 per cent geraniol, no adduct is formed. This happens in spite of the fact that there is a large excess of calcium chloride present and the contents while being stirred

TABLE 3—INFLUENCE OF RATIOS OF CALCIUM CHLORIDE TO OIL AND OIL TO SOLVENT ON YIELD OF GERANIOL (SOLVENT METHOD)

(Geraniol content of oil, 70% ; weight of oil used, 20 g. ; temperature of reaction, 30°C. ; time of stirring, 2 hr.)

No.	Calcium chloride : oil	Solvent : oil	Yield of geraniol %
1	3.75 : 1	5 : 1	49.3
2	2.5 : 1	2.5 : 1	48.5
3	1.5 : 1	2.5 : 1	45.0
4	0.75 : 1	2.5 : 1	31.5
1	2.5 : 1	2.5 : 1	48.5
2	2.5 : 1	5 : 1	48.2
3	2.5 : 1	10 : 1	46.1

TABLE 4—INFLUENCE OF GERANIOL CONTENT OF OIL ON YIELD OF GERANIOL (SOLVENT METHOD)

(Weight of calcium chloride used, 50 g. ; temperature of reaction, 30°C. ; time of stirring, 2 hr.)

No.	Wt. of oil g.	Geraniol in oil %	Yield of geraniol %	Geraniol in residue %	Recovery of geraniol %
1	25	40.0	. .	40.0	. .
2	25	44.8	12.3	29.4	26
3	20	50.2	20.5	31.3	38
4	20	60.6	30.4	37.0	50
5	20	73.1	50.0	38.0	63
6	20	80.1	60.0	52.0	73
7*	19.4	97.0	72.0	97.0	72
8	9.7	97.0	66.0	97.0	66

* In experiments Nos. 1 to 6 and in No. 8, 100 cc. petroleum ether was used. In the 7th experiment 150 cc. solvent was required to keep the mass fluid enough for stirring.

are quite fluid. It appears that the non-geraniol portion interferes with the adduct formation. It is also seen that the yield of geraniol increases by increasing the geraniol content or by diminishing the non-geraniol portion of the oil. The role of the solvent, besides facilitating good stirring and thereby hastening the reaction, is to moderate the retarding effect of the non-geraniol portion to a considerable extent, in spite of the fact that the solvent itself has its own retarding effect.

The effects of other solvents such as *n*-heptane, benzene, carbon tetrachloride, ethyl ether and acetone were also studied. It was found that no adduct formation took place in these solvents, except in *n*-heptane which had the same effect as the petroleum ether (b.p. 65-68°C.).

These observations show that the non-geraniol portion, whether present in the oil or added as solvent, retards the adduct formation. The terpenes (non-geraniol portion already present in the oil) and solvents like benzene and carbon tetrachloride, are more polar than solvents like *n*-hexane and *n*-heptane. The polar solvents appear to influence adduct formation by their dissociating influence on the adduct. They set up an equilibrium between the formation and dissociation of the adduct. This seems to be the plausible explanation for the incomplete adduct formation even when the original oil contains as high as 97 per cent free geraniol (Table 4, experiments 7 and 8).

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Chemical Examination of the Essential Oil from *Eugenia jambolana* Leaves

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The essential oil from *Eugenia jambolana* leaves has been examined. It appears to contain terpenes, *c.* 20 ; sesquiterpenes of the cadalene type, *c.* 40 and sesquiterpenes of the azulene type, below 10 per cent. Aldehydes, ketones and phenols have been found to be absent.

Eugenia jambolana or *jamun* in Hindi is an evergreen tree found all over India. The ripe berries are edible. The seeds contain an essential oil which was examined many years ago by Power and Callan[1]. The essential oil from the leaves has not so far attracted any attention. Preliminary work on this oil revealed that the oil has an agreeable odour and can be used in the compounding of synthetic oils and flavours. It was, therefore, considered worthwhile to investigate its chemical composition.

The yield of oil varies according to the season and was found to be higher in February than in July. There was also an appreciable variation in the physico-chemical properties. The oil has a tendency to polymerize rapidly on keeping.

EXPERIMENTAL

The leaves were collected from local sources and distilled in a steam-heated copper still. The condensate was absorbed in purified benzene kept in the receiver. The solvent was subsequently distilled off and the last traces removed under vacuum. The oil was dried with anhydrous sodium sulphate and filtered. The yield of the oil from the leaves collected in July-August was 0.016 per cent and from those obtained in January-February, 0.018 per cent. Aldehydes and ketones (bisulphite method), and phenols were found to be absent. The physico-chemical properties of the samples are given in Table 1.

Fractionation

Sixty grams of the oil (distilled in Jan.-Feb.) were fractionated under reduced pressure. Three main fractions were collected as given in Table 2.

TABLE 1—CHARACTERISTICS OF E. JAMBOLANA LEAF OIL

PARTICULARS	OIL DISTILLED IN JULY-AUG.	OIL DISTILLED IN JAN.-FEB.
Sp. gr.$^{20°}$	0.8986	0.8943
$n^{20°}$	1.4943	1.4999
$[\alpha]_D$ (in $CHCl_3$)	−20.9°	−13.2°
Solubility in 90% alcohol, vol.	1 in 5	1 in 5
Acid value	1.43	1.05
Ester value	25.34	35.47
Ester value after acetylation	93.1	66.89

TABLE 2—FRACTIONATION OF E. JAMBOLANA LEAF OIL

FRACTION	PRESSURE *mm.*	TEMP. °*C.*	WT. OF FRACTION *g.*	$n^{30°}$
I	10	Below 100	7.06	1.485
II	10	100-125	28.86	1.504
III	10	125-128	7.07	1.509
Residue & loss	10	. .	17.01	. .

TABLE 3—REFRACTIONATION OF FRACTION II

FRACTION	TEMP. °*C.*	PRESSURE *mm.*	WT. OF FRACTION	SP. GR.$^{35°}$	$n^{32°}$
a	Below 80	2	1.51	0.8884	1.485
b	80-85	2	1.59	0.8952	1.493
c	85-90	2	3.50	0.8999	1.496
d	Above 90	2	3.62	0.9018	1.504
Residue			0.78		1.509

Fraction I—This fraction, distilled twice over sodium metal, was a colourless mobile liquid with a characteristic smell resembling that of unripe mangoes. At ordinary pressure it boiled at 174-176°C. and showed $[\alpha]_D$ -13.1° and $n^{20°}$ 1.4740.

From its odour and physico-chemical properties, this fraction appeared to be a mixture of *l*-limonene and dipentene. On exposure to light and air, it underwent polymerization and discoloration, probably due to the presence of traces of highly unsaturated constituents. Attempts to prepare its crystalline derivatives were not successful.

Fraction II—This fraction (*c.* 20 g.) was distilled twice over sodium, when a mobile colourless liquid (*c.* 11 g.) with a faint odour was obtained. It had sp. gr.$^{30°}$ 0.9137, $n^{33°}$ 1.499 and $[\alpha]_D$ − 26.6°. It was again fractionated (Table 3).

The estimation of unsaturation by titration against perbenzoic acid showed that the above fractions contained mixtures of bicyclic and tricyclic sesquiterpenes.

Dehydrogenation—All the four sub-fractions of fraction II were dehydrogenated separately with sulphur in the usual manner. The product in each case was distilled over a pinch of zinc dust when a bluish green distillate was collected. This was dissolved in petroleum ether and extracted with phosphoric acid (85 per cent).

The phosphoric acid layer after separation was washed with petroleum ether and decomposed with water at a low temperature. A greenish oil, showing the presence of azulenes was obtained.

The petroleum ether layer was extracted once again with phosphoric acid to remove residual azulenes, washed with water and dried. After the removal of the solvent a yellow oil was obtained.

The picrates of both these products were prepared. The yellow oil gave brilliant orange coloured needles which through their mixed melting point (115°C.) were identified as those of cadalene picrate. The azulenic portion gave a black picrate melting at 115-116°C. and is probably identical with S-guaiazulene (m.p. 121°C).

Fraction III—This fraction was also dehydrogenated with sulphur. The constituents were found to be the same as those of fraction II, with lesser proportions of cadalene and greater of azulene.

The oil thus appears to contain terpenes, *c.* 20 ; sesquiterpenes of the cadalene type, *c.* 40 and sesquiterpenes of the azulene type, below 10 per cent.

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Examination of the Essential Oil from *Siyah Zeera (Carum bulbocastanum)*

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The seeds of Indian *siyah zeera* are considered to be derived from *Carum carvi* by some authors and from *Carum bulbocastanum* by others. The essential oil from the seeds of *siyah zeera* obtained from Peshawar and identified as those of *Carum bulbocastanum*, has been examined.

Refractive index/density curve method was used and it has been concluded that the essential oil may contain cuminic alcohol, car-4-ene, α-terpinene and β-terpinene.

The seeds of *siyah zeera* are dark brown to black in colour, possess an aromatic spicy taste and are extensively used in condiments. They are the dried fruits of the plant, about 2.0 to 5.4 mm. in length, 0.6 to 1.2 mm. in breadth and 0.5 to 0.8 mm. in thickness.

The *siyah zeera* is an umbelliferous plant cultivated in India. It also grows wild in the regions of Afghanistan, Kashmir, Hindukush and Peshawar at an altitude of 7,000 to 10,000 ft. The seeds sold in the Indian market under this name, however, do not seem to come from a single botanical species. They are said to be derived from *Carum carvi*[1] Linn. and *Carum bulbocastanum*[2] W. Koch by different authors. They have also been called *Carum indicum* Regel & Herd. and *Carum nigram* W. Royle.

The authors carried out investigations on the seeds specially collected from plants grown near Peshawar. The seeds were sent for identification to prominent botanists in India and abroad. In their opinion they were derived from *Carum bulbocastanum*.

One of us (R.A. Dhaul[3]), examined the essential oil obtained from the seeds and found that it contained cuminic aldehyde ; terpinolene, ocimene, *p*-cymene and carvone were absent. The presence of cuminic aldehyde was confirmed by preparing its semicarbazone.

As this finding showed a great variation in the composition of the oil from that reported by Nadkarni[4] (carvone 45-60 per cent, *d*-limonene and cymene), the work was continued and attempts were made to identify the constituents of the oil by the more modern refractive index/density graph method recommended by Sutherland[5] and Murray[6]. In the graph it is found that (*i*) intermediate fractions are spread out ; (*ii*) similar fractions fall together ; and (*iii*) pure substances appear at the angles of the curve. According to

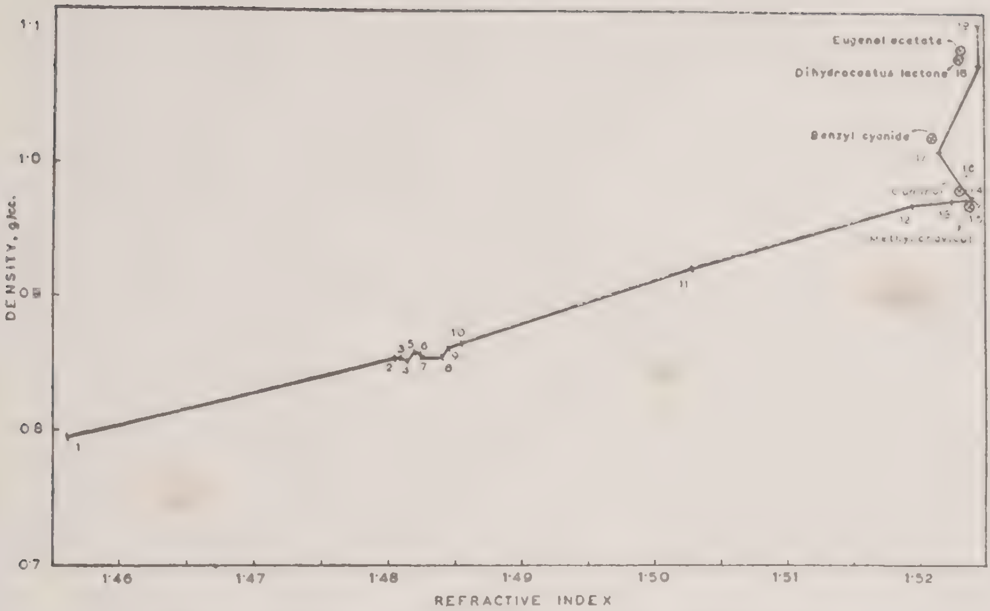

FIG. 1—REFRACTIVE INDEX/DENSITY GRAPH FOR ESSENTIAL OIL FROM *Carum bulbocastanum*

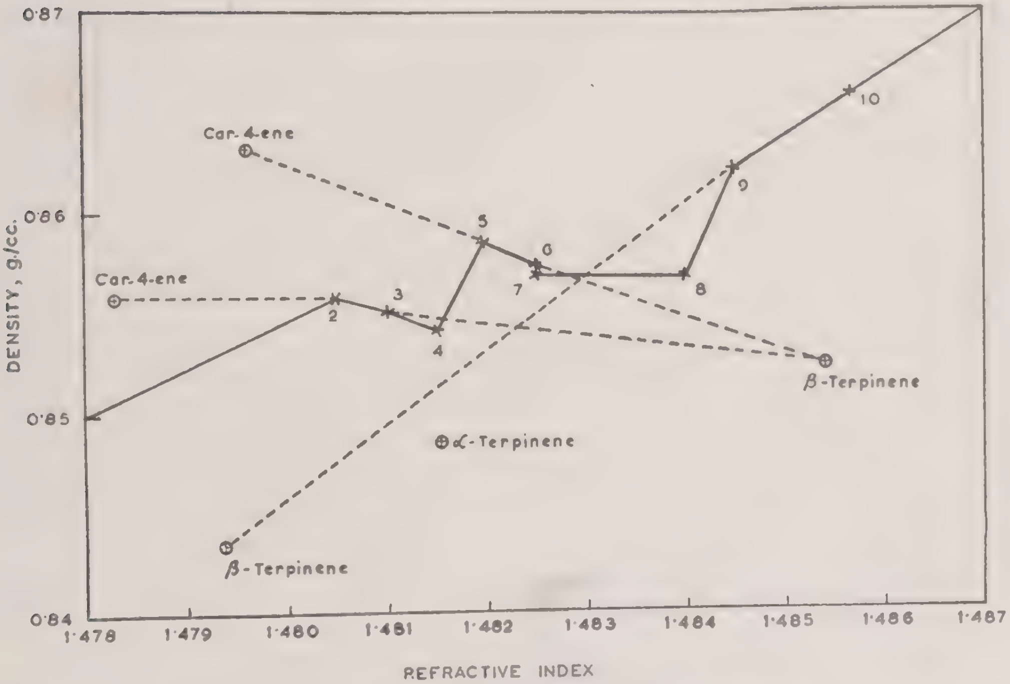

FIG. 2—A PORTION OF GRAPH (FIG. 1) REPRESENTING TERPENE FRACTIONS MAGNIFIED

Murray[6] the particular value of this representation lies in the fact that it is often possible to fix fairly accurately the physical constants of individual compounds present (as mixtures) in impure fractions.

The essential oil (250 cc.) was fractionally distilled and 19 fractions of 10 cc. each were collected. The refractive indices and the specific gravities of these fractions were determined within 36 hours of the distillation at 15°C.

A curve was obtained by joining, in order, points representing each successive fraction by straight lines. For ready reference, the points representing the densities and the refractive indices of pure substances likely to be present were also shown in the graph.

Results and Discussion

By extrapolation and other considerations it was concluded that: fraction 1 may be a mixture of a hydrocarbon or hydrocarbons with terpenes ; fractions 2, 3 and 4 may be mixtures of car-4-ene, β-terpinene and α-terpinene ; fractions 5, 6 and 7 may be mixtures of car-4-ene and β-terpinene ; fractions 8, 9 and 10 may contain a great proportion of β-terpinene ; fractions 11 and 12 may be considered as mixtures mainly of β-terpinene and cuminic alcohol ; fractions 13, 14 and 15 may contain cuminic alcohol apparently with some quantity of methyl chavicol ; fraction 16 may be cuminic alcohol ; fraction 17 may probably contain benzylcyanide ; while fraction 18 may contain compounds like eugenol acetate and dihydrocostuslactone.

The above results indicate the absence of carvone in the essential oil. The presence either of *d*-limonene or cymene is also not indicated. These results agree with Dhaul's observations[5]. They, however, do not indicate the presence of cuminic aldehyde detected and confirmed by Dhaul[5]. It must, therefore, be present in small quantities insufficient to influence the constants of any fraction.

It may be tentatively concluded that the essential oil from the seeds of *Carum bulbocastanum* contain cuminic alcohol, car-4-ene, β-terpinene, and α-terpinene.

Since these results are not in agreement with the findings of previous workers, study of the chemical constituents of the oil is being continued.

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Chemical Examination of Dill Seed and Herb Oils

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Indian dill herb oil and dill seed oil have been chemically examined and their characteristics have been compared with oils of foreign origin.

Anethum graveolens Linn., commonly known as dill, is cultivated in the United States of America and European countries. Indian dill or *Anethum sowa* Roxb. Syn. *Peucedanum graveolens*[1] Benth., differs from foreign dill in chemical characteristics. It is called Persil-de-Marais[2] in French, *satpushpi* or *misariya* in Sanskrit ; *sowa* or *soya* in Hindi, Bengali and Punjabi ; *satakuppi* in Tamil ; *surva* in Gujarati and *shepu* in Marathi.

The herb is cultivated throughout India[3]. Its fruits or seeds, which are very light (about 25,000 to an ounce), are used as carminative, stomachic, stimulant and diuretic, while dill water is administered in flatulence, hiccup, colic, and abdominal pains.

Young leaves and leaves from young plants are more aromatic than older leaves and leaves from mature plants. They are widely used as a flavouring agent. Dill vinegar is prepared by soaking the seeds in vinegar for a few days. In France[4] dill seeds are used for flavouring preserves, cakes, pastries and sauces, and in Germany for pickling cucumbers.

Dill grows well in a light medium sandy loam. The details of its cultivation are given by Guenther[5]. The herb is susceptible to strong winds and heavy rains. High temperature reduces the yield of the oil considerably.

In case the herb oil is to be prepared, the crop should be harvested at the stage when the flavour of the isolated oil is identical with that of the herb. If, however, the seeds are required, the harvest should be reaped when the maximum number of seeds are ripe. The harvesting should be done early in the morning or late in the evening. To prevent loss of seeds care should be taken to handle the plants with the least possible shaking. Under favourable conditions, the yield of dill seeds may amount to 500-700 lb. per acre.

DILL HERB OIL

For the preparation of dill herb oil, fresh tender herb is distilled in steam for 2 to 4 hours. More mature herb requires more time for complete distillation, which may take 8 to 10 hours. The oil obtained from the mature herb is richer in carvone and resembles dill seed oil.

Depending on the maturity of the herb the properties of dill herb oils vary widely. For flavouring purposes, herb oils with low ketone (carvone) content are preferred.

The typical flavour of the herb oil is due to *d*-phellandrene ; the higher the proportion of this terpene the more the oil resembles the fresh herb. The oils containing 20 per cent or less of carvone have the finest and more pronounced herb character. This character predominates so long as the carvone content of the oil does not exceed 35 per cent.

Malviya & Dutta[6] have examined the Indian dill herb oil. Its characteristics along with those of American and Hungarian dill herb oils are given in Table 1.

Commercial dill herb oil is frequently adulterated with limonene and other terpenes resulting from the preparation of sweet orange oil concentrates. The most common adulterants are the terpenes obtained in the extraction of carvone from caraway seed oil. The detection of these adulterants is not possible through ordinary routine analyses.

DILL FRUIT OIL

The dill seed is crushed prior to distillation between heavy rollers. The crushed seed is mixed with water and distilled in steam for 10 to 12 hours. The yield of the oil varies from 1.3 to 3.5 per cent.

Dill seed oil is pale yellow in colour. Rao *et al.*[7] and Malviya & Dutta[6] have examined the Indian dill seed oil. The characteristics of these oils are given in Table 2.

It will be noticed that the specific gravity of the Indian dill seed oil is much higher than that of the other oils. According to the British Pharmacopoeia dill seed oil should have: sp. gr.$^{20°}$, 0.895-0.910 ; $[\alpha]_D$, +70° to +80° ; $n^{20°}$, 1.431 to 1.492 and carvone content, not less than 43 per cent and not more than 63 per cent. It should be soluble in equal volume of 90 per cent alcohol and 10 volumes of 80 per cent alcohol.

Malaviya & Dutta[6] have stated that the exhausted fruits or seeds contain 16.3 per cent fat and 15.1 per cent protein. The Indian dill seed oil has been fractionated into lighter and heavier than water fractions[6,7]. Their characteristics along with the whole Indian oil are given in Table 3.

Chemical composition—Dill herb oil has been found to contain *d*-phellandrene, dipentene, carvone, dillapiole, isomyristicin and myristicin. Elze[8] obtained a volatile oil containing n-octyl alcohol by distilling dill herb oil in steam.

Dill seed oil contains carvone, *d*-limonene, phellandrene and dillapiole, the last substance being found mainly in Indian seed oils though its presence has also been detected[9] in European oils. Branigan[10] has reported the presence of α-pinene, dipentene and dihydrocarvone in the oil. The presence of dihydrocarvone, in Indian dill seed oils has been noted by Bhattacharyya & Chakravarti[11].

TABLE 1—CHARACTERISTICS OF AMERICAN AND HUNGARIAN DILL HERB OILS FROM ANETHUM GRAVEOLENS LINN. AND INDIAN DILL HERB OIL FROM PEUCEDANUM GRAVEOLENS BENTH.

	AMERICAN[5] OIL	HUNGARIAN[5] OIL	INDIAN[8] OIL
Sp. gr.$^{15^\circ}$	0.880-0.910	0.897-0.908	0.8726 at 20°C.
$[\alpha]_D$	+86° to +105°	+81°10′ to +101°4′	+286.6°
n^{20°	1.4770 to 1.4840	1.4800 to 1.4855	1.487
Carvone content, %	12.0-40.0	20.6-42.5	..
d-phellandrene, %	..	..	74.6
Acid val.	..	..	2.10
Ester val.	..	..	6.72
Solubility, in vol. alc., %	0.5-1.5 or more of 90 ; in usual	in 80, in some cases clear, in others cloudy occasionally opalescent	

TABLE 2—CHARACTERISTICS OF HUNGARIAN, ENGLISH, GERMAN AND RUSSIAN DILL SEED OILS FROM ANETHUM GRAVEOLENS LINN. AND THE INDIAN DILL SEED OIL FROM PEUCEDANUM GRAVEOLENS BENTH.

	HUNGARIAN[5] OIL	ENGLISH[5] OIL	GERMAN[5] OIL	INDIAN[5] OIL	RUSSIAN[5] OIL
Sp. gr.$^{15^\circ}$	0.895-0.915	0.898-0.915	0.9002	0.9785	0.918
n^{20°	1.484-1.491	1.483-1.488	..	1.4943	1.4882
$[\alpha]_D$	+70° to +82°	+74° to +82°	+70.25°	+47.6°	+71°10′
Ketones as carvone, %	40-60	48-51	..	19.5	53.6
Solubility in vol. alc., %	4 to 9 of 80 ; exceptionally in 1.5 to 2.0	7-10 of 80	..	..	1 to 1.5 of 80
Oil on wt. of the fruit, %	2.3-3.5	4.0	3.8	3.19	..

TABLE 3—CHARACTERISTICS OF INDIAN DILL SEED OIL AND ITS (LIGHTER AND HEAVIER THAN WATER) FRACTIONS

	RAO *et al.*[7]			MALAVIYA & DUTTA[6]	
	Whole oil	Lighter oil	Heavier oil	Lighter oil	Heavier oil
Sp. gr.$^{15^\circ}$	0.9785	0.9313	1.0935	0.9719 at 20°C.	1.0573
n^{20°	1.4943	1.4853	1.5134	1.4905	1.5385
$[\alpha]_D$	+47.6°	+58.2°	..	+38.5°	+23.6°
Ketones as carvone, %	19.5	17	..	..	..

TABLE 4—PHYSICO-CHEMICAL PROPERTIES OF INDIAN DILL OILS

	HERB OIL	SEED OIL
Sp. gr.$^{30°}$	0.9241	0.9341
$n^{30°}$	1.4864	1.4854
$[\alpha]_D$	+71°48′	+54°
Acid val.	1.9	1.2
Ester val.	43.3	37.7
Ester val. after acetylation	82.5	56.3
Carvone (by neutral sulphite method), %	13.5	41.7
Phenols (by caustic potash method), %	5.6	. .

TABLE 5—SOME CHARACTERISTICS OF FRACTIONS OF INDIAN DILL HERB OIL

FRACTION NO.	BOILING POINT °C.	WEIGHT g.	FRACTION OF THE OIL %
1	61-80	16.05	9.72
2	80-84	35.32	21.60
3	84-90	18.93	11.19
4	90-96	15.25	9.28
5	96-114	6.77	4.09
6	114-124	13.42	8.13
7	124-138	4.13	2.50
8	138-165	10.28	6.21
9	165-175	28.63	17.36
10	Residue	11.86	17.18
	Loss	4.36	2.68
		165.00	100.00

The present work was undertaken with a view to the commercial utilization of the Indian dill oils. The oils have been fractionally distilled and these characteristics studied.

EXPERIMENTAL

The herb and seed oils were prepared by steam distillation of the green herb and mature fruits. The yields of oils were 0.4 and 2.5 per cent respectively. The physico-chemical properties of the herb and seed oils are given in Table 4.

DILL HERB OIL

The oil (165 g.) was fractionated at 25-30 mm. The descriptions of the fractions collected are given in Table 5.

The physico-chemical constants of the fractions given in Table 5 have been compiled in Table 6.

Fractions 1, 2, 3 and 4 were combined together in the proportions in which they were obtained and redistilled over metallic sodium under reduced pressure. The percentage of terpenes in the combined fractions was 86.9 per cent ; (44.5 per cent on weight of whole oil).

The presence of d-α-phellandrene in the combined fraction was determined by preparing its nitrosite which melted at 118-119°C. after recrystallization. The presence of limonene was not apparent as the fraction did not yield a crystalline tetrabromide derivative.

Fractions 4, 5, 6 and 7 contained 45.21, 57.6 and 59.8 per cent of carvone (neutral sulphite method) respectively. The presence of d-carvone was confirmed by its semicarbazone which melted at 162-163°C.

Fractions 8 and 9 were mixed together in the proportions in which they were obtained and freed of their carvone content. The residual oil was treated with 4 per cent sodium hydroxide solution. The undissolved non-phenolic portion was separated. The phenolic fraction was liberated from the alkaline solution by acidifying with dilute sulphuric acid. The oily layer was extracted with ether. The extract was washed free of acid dried and the ether distilled off. The phenolic fraction gave a benzoyl derivative melting at 69-70°C. From these studies the Indian dill herb oil appears to contain, terpenes, chiefly d-α-phellandrene, 44.57 ; d-carvone, 13.5 ; alcohols, calculated as $C_{10}H_{18}O$, 11.1 ; esters, as linalyl acetate, 15.05 and phenols, 5.6 per cent. Eugenol may be present.

DILL SEED OIL

The oil (173 g.) was fractionated at 30-35 mm. The fractions collected are given in Table 7. These fractions were analysed individually. Their physico-chemical constants are given in Table 8.

Fractions 1, 2 and 3 were mixed together in the proportion in which they were obtained and re-fractionated over metallic sodium. The terpenes in these fractions were 65.05 per cent (28.77 per cent on the weight of original oil). The presence of limonene in the terpenes was confirmed through its tetrabromide derivative which melted at 103-104°C. The mixed fraction did not give a positive nitrosite test, thereby indicating the absence of phellandrene.

Fractions 4, 5, and 6 also, were mixed in the proportions in which they were obtained from the oil. The carvone isolated from combined fraction gave a semicarbazone identical to that of d-carvone (m.p. 162-163°C). The carvone free fractions were re-fractionated at 6-8 mm. The fraction boiling at 88-90°C. showed $[\alpha]_D^{30}$, $-13°12'$ and n^{30}, 1.4680. It was identified as dihydrocarvone through its semicarbazone derivative (m.p. 188-189°C.) ; its yield on the weight of the oil being 7.2 per cent. The fraction boiling at 152-155°C. at 8 mm. (15.6 per cent on weight of oil) was taken to be dillapiole and could not be further examined.

TABLE 6—PHYSICO-CHEMICAL CONSTANTS OF THE VARIOUS FRACTIONS OF INDIAN DILL HERB OIL

Fr.	Sp. gr.$^{30°}$	$n^{30°}$	$[\alpha]_D$	Acid val.	Ester val.		Carvone content %
					Initial	After acetylation	
1	0.8393	1.4675	122°54′	Nil	34.09	. .	. .
2	0.8395	1.4670	123°42′	Nil	38.67	. .	2.04
3	0.8452	1.4685	112°18′	0.2	38.54	. .	3.56
4	0.8777	1.4775	72°48′	0.44	44.9	. .	4.7
5	0.9416	1.4825	29°24′	1.16	52.35	. .	45.21
6	0.9632	1.4880	28°48′	1.47	55.99	. .	57.62
7	0.9867	1.4940	. .	1.58	56.1	. .	59.83
8	1.0418	1.5055	12°30′	1.84	. .	. .	40.03
9	1.0912	1.5195	4°18′	2.3	49.4	52.4	5.04
10	1.1010	1.5200	. .	9.7	72.29	. .	. .

TABLE 7—SOME CHARACTERISTICS OF INDIAN DILL SEED OIL

Fraction No.	Boiling point °C.	Weight g.	Yield on weight of oil %
1	70-78	14.5	8.38
2	78-90	46.6	26.36
3	90-100	16.3	9.42
4	100-125	41.3	23.87
5	125-140	19.2	11.09
6	140-170	24.7	14.27
7	Residue	8.0	4.62
	Loss	2.4	1.99
		173.0	100.00

TABLE 8—PHYSICO-CHEMICAL CONSTANTS OF VARIOUS FRACTIONS OF INDIAN DILL SEED OIL

Fraction No.	Sp. gr.$^{15°}$	$n^{30°}$	$[\alpha]_D$	Acid val.	Ester val.		Carvone content %
					Initial	After acetylation	
1	0.8543	1.4690	92°24′	Nil	15.6	. .	7.07
2	0.8608	1.4700	88°	Nil	16.1	. .	12.8
3	0.8969	1.4750	62°48′	Nil	22.5	58.4	38.7
4	0.9581	1.4860	52°12′	0.83	40.56	. .	91.2
5	0.9900	1.4950	35°24′	1.04	44.4	75.2	67.5
6	1.1075	1.5170	12°48′	2.2	44.9	. .	20.4
7	. .	1.5245	. .	. .	. .	. .	6.7

The Indian dill seed oil, according to these studies, appears to contain, terpenes, chiefly limonene, 28.77 ; d-carvone, 41.7 ; dihydrocarvone, 7.2 and dillapiole 15.6 per cent.

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Chemical Examination of Some Less Familiar Essential Oils

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The essential oils of the medicinal plants *Zataria multiflora* Boiss., *Seseli indicum* W. & A., and *Apium graveolens* Linn. have been examined. The oil from the first plant has been found to contain carvacrol, 69 ; *p*-cymene, 17 ; borneol, 3 ; zatarol, 0.5 ; zatarinol, 2 ; and an acid, 0.6 per cent. The oil from the seeds of the second consists of *c*. 47 per cent of *d*-limonene along with selinene, isovaleric acid, seselin, bergaptene, two acids and traces of a phenol. The oil from the seeds of the last plant contains *d*-limonene, *c*. 44 ; and selinene, *c*. 23 per cent. Its physico-chemical properties agree well with those of the oils obtained in European countries.

Essential oils play quite an important part in medicine. The oils of eucalyptus and cajuput, which carry large proportions of cineol, are used in the treatment of chronic bronchitis and rheumatism, and are useful stimulants. Thymol possesses bactericidal properties, camphor is a heart stimulant and carvacrol is an antiseptic and germicide. Some essential oil bearing drugs are used in the crude form as they are available in the market. *Zataria multiflora* Boiss., *Seseli indicum* Wight & Arnold and *Apium graveolens* Linn., whose essential oils form the subject of study of this paper, are three such drugs.

Zataria multiflora Boiss.

Zataria multiflora Boiss. (*Saatare-farsi*) is a small herbaceous plant belonging to the family Labiatae[1]. The dried plant is used in India in infusion as an agreeable aromatic stimulant and diaphoretic, and also for other ailments. It is sold in the bazaar under the name of 'Saatar'. It has a fragrant odour and consists of small leaves mixed with portions of slender woody stems and numerous minute flowers[2]. Dymock[2] mentions the occurrence of an aromatic essential oil in its leaves.

The fresh drug consisting mostly of leaves mixed with portions of stems and flowers (as is usually available and used) was obtained from the market in air-dried condition. Steam distillation of the powdered drug (moisture content, 12.8 per cent) yielded 0.6 per cent of a golden-yellow essential oil[3] having a characteristic odour, a bitter and sharp taste and a biting effect on sensitive parts of the skin. It is rich in carvacrol and possesses the following characteristics: d^{32}_{32}, 0.93098 ; $n_D^{32°}$, 1.5114 ; $[\alpha]_D^{32°}$, $-0°53'$; acid value, 1.3 ; saponification, 8.0 ; and saponification value after acetylation, 175.9.

The oil is soluble in 2.5-3 vols. of 70 per cent alcohol and boils off between 172° and 240°C. leaving behind a brownish red viscous residue.

The oil has been found to contain total phenols (69 per cent) made up mainly of carvacrol with traces of thymol. The neutral portion (31 per cent) besides containing borneol (3 per cent) is rich in *p*-cymene (17 per cent). Along with the above constituents have been found: (*i*) an optically inactive phenol, which is a deep yellow thick liquid with a creosote-like odour (b.p., 234-35°/746 mm. ; d^{28}_{28}, 0.98112 ; $n_D^{28°}$, 1.51774 ; yield, 0.5 per cent), insoluble in water and soluble in 3.5-4 vols. of 65 per cent alcohol. Its alcoholic solution gives a deep violet colour with ferric chloride. It yields 3:5-dinitrobenzoate melting at 93-94°C. It appears to be a new phenol and therefore has been tentatively named "zatarol" ; (*ii*) an alcohol ($C_{10}H_{16}O$), which is a colourless oily liquid (b.p., 205-206°C. ; yield, 2.0 per cent) tentatively called "zatarinol" ; and (*iii*) a pale yellow liquid acid (b.p., 209-211°C. ; n^{25}, 1.4888 ; yield, 0.6 per cent).

Seseli indicum Wight & Arnold

Seseli indicum Wight & Arnold (Sans. Vanayamani), belongs to the Natural Order Umbelliferae. It is an annual, erect or diffuse herb having white or pinkish flowers. The fruits (seeds) are often pale yellow[4]. The seeds act as an anthelmintic for roundworms, are administered as stimulant, carminative and stomachic, and are also used for other purposes. In the market they are sold under the misnomer '*Ajmoda*'. The fresh seeds were obtained locally and identified by the authorities of the Indian Botanic Garden, Sibpur, Calcutta, as those of *Seseli indicum* Wight & Arnold.

Three crystalline coumarins, seselin, bergaptene and isopimpinellin were obtained by Bose and Guha[5] and Spath *et al.*[6] through petrol extraction of the seeds. The limpid and almost colourless essential oil obtained in these laboratories from the air dried and powdered seeds by steam distillation (average yield, 2.5 per cent), develops a slightly yellowish shade on keeping or heating. It has a characteristic aromatic odour resembling that of the seeds and possesses a clove-like taste. It showed d^{32}_{32}, 0.8905 ; $[\alpha]_D^{32}$, +34.7 ; n_D^{32}, 1.48214 ; acid value, 3.1 ; ester value, 30.0 ; saponification value, 33.1 ; and saponification value after acetylation, 64.6. It is soluble in 3.5 vols. of 90 per cent alcohol. The total quantity of alcohols calculated as $C_{10}H_{18}O$ was 17.13 per cent. Practically half of the oil distilled between 174-195°C. and of the remainder a large portion distilled above 250°C.

The oil on treatment with sodium bicarbonate solution (3 per cent) yielded a mixture of three acids (about 2 per cent). One of the acids has been found to be isovaleric acid ; the second boils at 227-231°C, and the last melts at 110-111°C. and is identical with the acid obtained from the oil through its hydrolysis by alcoholic potash. Caustic soda (5 per cent solution) extract of the oil (yield, 1.5 per cent) contained seselin (m.p. 120-121°C.) bergaptene (m.p. 189-190°C.) and traces of a phenolic substance. On hydrolysis with alcoholic caustic potash, the oil gave a reddish brown semi-solid mass (4 per cent) from which colourless shining crystals of an acid (m.p. 112-114°C.) have been isolated. The low boiling fractions of the oil (47 per cent) are made up mainly of *d*-limonene, while a major portion of the high boiling part of the oil seems to contain selinene.

Apium graveolens Linn.

Apium graveolens Linn. (Karaffs, Eng.-celery), belongs to the Umbelliferae family. The seeds are utilized medicinally as a diuretic, pectoral, and carminative adjunct to purgatives[7]. Celery is a popular European domestic remedy for the treatment of rheumatism[8]. Celery oil is known to be used for several purposes, but the chemical composition of the oil from purely Indian variety does not seem to have been investigated, though a few physical constants of the oil obtained from celery seeds imported from India have been determined by Messrs *Fritzsche Bros.*, New York[9].

The air-dried karaffs seeds were obtained locally. On steam distillation they yielded a slightly pale yellow thin oil having a characteristic smell resembling that of the seeds. The physico-chemical properties of the oil were determined and are given in Table 1 along with the comparative data recorded in literature ; they agree well with the values given by Gildemeister and Hoffmann[10].

The essential oil on extraction with caustic soda solution (5 per cent) gave a thick syrupy mass (yield, 1.42 per cent) which gave a colourless product melting at 61-63°C. On hydrolysis of the oil with alcoholic potash a semi-solid mass (10.7 per cent) was obtained. This was further divided by sodium

TABLE 1—COMPARATIVE CHARACTERISTICS OF THE ESSENTIAL OIL FROM A. GRAVEOLENS LINN.

CHARACTERISTICS	AUTHOR'S SAMPLE	GILDEMEISTER & HOFFMANN[10]	FRITZSCHE BROS.*
Sp. gr.	0.8849 at 20°	0.866-0.898 at 15°	0.872-0.891 at 15°
n^{20}	1.4848	1.478-1.486	1.4803-1.4841
$[\alpha]_D$	+60°, 47′ at 20°	+51°, 0′, to +82°, 0′	+65°, 53′ to +76°, 51′
Acid number	4.3	up to 4.0	..
Ester number	56.46	16.0-55.0	25.1-47.6
Ester number after acetylation	67.0	43.0-67.0	..
Solubility, in vol.	9.5-10 of 85% alc. ; 4.5 of 90% alc. (vol.)	6-8 of 90% alc. mostly turbid ; 1-1.5 of 95% alc. (vol.) ; sometimes opalescent with more	turbid in 90% alc. (vol.)

* From imported Indian seeds

carbonate solution (10 per cent) into (*i*) sodium carbonate soluble portion (6.0 per cent), probably acids ; and (*ii*) sodium carbonate insoluble portion (4.68 per cent). The low boiling portion of the oil shows mainly the presence of *d*-limonene (about 44 per cent) mixed up with some dipentene. One of the high boiling fractions of the oil indicates the presence of selinene. The occurrence of limonene to the extent of about 60 per cent and selinene about 10 to 15 per cent has been reported. In the present sample the terpene is estimated to be about 44 per cent and the sesquiterpene about 23 per cent.

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Hydration of Longifolene

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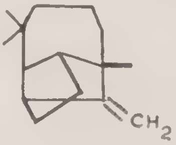
Longifolene, on hydration with acetic acid-sulphuric acid in dioxane, gave longiborneol and other alcohols besides a strongly laevorotatory hydrocarbon.

Recently Naffa and Ourisson[1] have proposed the structure (I) for longifolene, the sesquiterpene hydrocarbon from the essential oil of *Pinus longifolia*. It is based on new chemical evidence and the X-ray study of longifolene hydrochloride by Moffett and Rogers[2].



I

The present study in the hydration of longifolene has been conducted with a view to further elucidate its chemistry and, also, to arrive at precursors suitable for dehydrogenation to an azulene.

Hydration of longifolene—The sample of longifolene used in this study boiled at 118-119°C./mm. and showed d_4^{26} 0.9319, n_D^{26} 1.5015, M_D 64.71 (calculated for $C_{15}H_{24}$ 64.45), $[\alpha]_D + 46.26°$ (homogeneous), $+46.98°$ (in 5.326 per cent alcoholic soln).

The sesquiterpene, when treated with acetic acid-sulphuric acid in dioxane gives a mixture of acetates (29.9 per cent) and an isomerised hydrocarbon, (62.5 per cent), which is strongly laevorotatory $[\alpha] = -80.4°$. The hydrolysis of the ester mixture followed by fractional distillation yielded a colourless viscous liquid (88.6 per cent) and a white waxy crystalline solid (A) (9.8 per cent).

Solid alcohol (A)—It crystallises from petroleum ether in white flakes melting at 107-108°C. Its 3:5-dinitrobenzoate melts at 144-145°C.

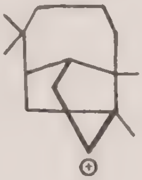
Isomerised hydrocarbon—It is a colourless mobile liquid, apparently homogeneous. It boils at 115-116°C./9.5 mm. and shows n_D^{26} 1.4985, $[\alpha]_D = -80.4°$ (homogeneous).

On preliminary oxidation with chromic acid-acetic acid it gave a ketonic product which forms a red 2:4-dinitrophenylhydrazone melting at 200-202°C. and a semicarbazone which melted at 232° (decomp.).

Reactive ketone—The colourless viscous liquid alcohol obtained from the hydrolysate of the ester mixture boils at 147-149°C./8.5-9 mm. On oxidation with chromic acid it yielded a colourless mobile liquid boiling at 129°C./4 mm. and showing n_D^{24} 1.4990, n_D^{30} 1.4980, d_4^{30} 0.9902, RM_D 65.14 (calculated for $C_{15}H_{24}O$ 64.9) and $[\alpha]_D + 30.28°$ (in 3.302 per cent alcoholic soln). It gave a semicarbazone melting at 220-221°C. (decomp.). The 2,4-dinitrophenylhydrazone obtained from the semicarbazone crystallised in orange red needles from alcohol and melted at 156-158°C.

On reduction with sodium and propyl alcohol or with lithium aluminium hydride, a crystalline solid was obtained in quantitative yield. After repeated crystallisations from petroleum ether it was found to be identical with the solid alcohol (A) through their mixed melting point.

The alcohol can possibly result from the carbonium ions shown below:



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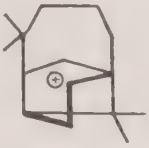

III

Hindered ketone—The colourless mobile liquid obtained after the separation of the semicarbazone from the chromic acid oxidation product boils at 100°/0.6 mm. and shows n_D^{26} 1.4990, d_4^{26} 0.9949, M_D 64.95 (calculated for $C_{15}H_{24}O$, 64.9) and $[\alpha]_D + 18.79°$ (in 5.32 per cent alcoholic soln). It does not give any carbonyl derivative.

On reduction with sodium and propyl alcohol it gave, in quantitative yield, a solid product, which after purification by vacuum sublimation was identified with longiborneol (m.p. 105-106°C.) through their mixed melting point. The 3:5-dinitrobenzoate of the alcohol crystallised in white needles from a mixture of benzene and petroleum ether and melted at 155-156°C.

When lithium aluminium hydride was used as a reducing agent a colourless viscous liquid, which does not yield a solid 3:5-dinitrobenzoate, was obtained.

Solid alcohol (B)—It was separated from hindered ketone through chromatographic isolation. It crystallises from *n*-hexane in white flakes melting at 92-94°C. It does not form a 3:5-dinitrobenzoate and is stable to oxidation with chromic acid even under drastic conditions. It appears to be a tertiary alcohol, probably by direct addition of H_2O without any rearrangement.

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Dihydrojasmone

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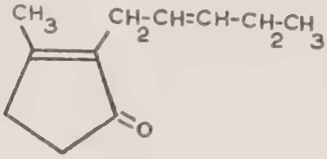
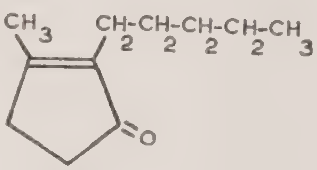
Dihydrojasmone has been prepared from γ-n-hexyl-γ-valerolactone in 92·95 per cent yield.

Jasmone (I) which forms about 3 per cent of the oil from the flowers of *Jasminum grandiflorum* was first isolated by Hesse[1] in 1899. The ketone also occurs to some extent in the oils of jouquille[2], orange blossoms[3], peppermint[4] and niroli water[3].

The structure of the ketone was independently established by Ruzicka and Peiffer[5], and Treff and Werner[6]. Its first synthesis was achieved by the latter workers[7] in 1935. Recently, natural jasmone has been shown[8] to possess the cis-configuration.



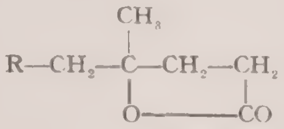
I



II

Jasmone appears to be a necessary component for producing a true jasmine note. Since commercial synthesis of (I) has not been worked out so far, a large number of related compounds[9] have been synthesised and their odours determined. It is significant that dihydrojasmone (II) shares the jasmine odour closely and is a valuable synthetic.

Of the various methods available for the synthesis of cyclopentanones, the intramolecular conversion of suitably constituted lactones to the corresponding ketones has certain attractive features. In 1938, Maschmeijer[10] described a procedure for the preparation of cyclopentanones from γ-lactones by the action of sulphuric acid. Frank and co-workers[11] converted the lactones into the unsaturated ketones by flash distillation with phosphorus pentoxide. However, the drawback of these methods is that the yield of the desired ketone is only 20 to 50 per cent. A new method[12], which can accomplish this conversion in yields of 92-97 per cent has been developed. The method consists in heating the lactones with polyphosphoric acid at 100°C. for 1½ to 2½ hours. When this procedure was applied to γ-n-hexyl-γ-valerolactone (III), dihydrojasmone (II) was obtained in a yield of 92 to 95 per cent.



(R = *n* = Am)

III

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Structure and Synthesis of Lactone of Tricycloekasantalic Acid—Degradation Product from Sandalwood Oil

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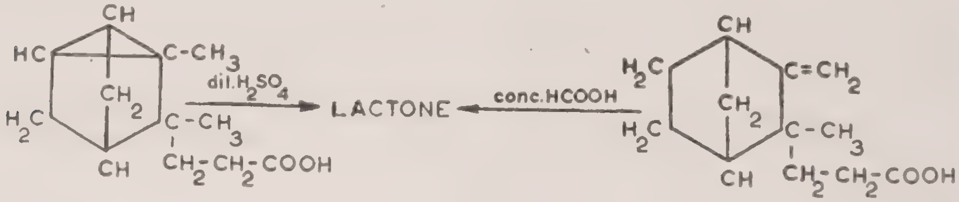
The structure of the lactone of tricycloekasantalic acid has been determined and confirmed by its synthesis. During the formation of the lactone from tricycloekasantalic (I), or bicycloekasantalic acid (II) a structural rearrangement takes place. The mechanism of the rearrangement has been discussed.

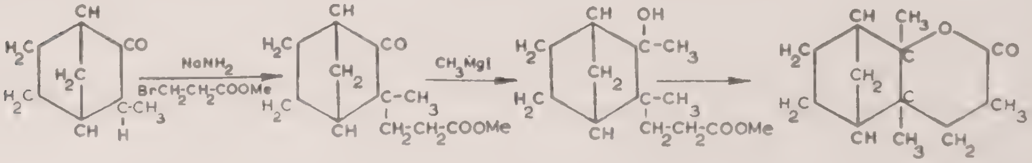
The lactone, $C_{12}H_{18}O_2$ (m.p. 102-103°C.), is prepared by refluxing tricycloekasantalic acid (I) with dilute sulphuric acid. It is also obtained[2] from bicycloekasantalic acid (II) by a similar treatment with concentrated formic acid.



I
Tricycloekasantalic acid

II
Bicycloekasantalic acid

Bhattacharyya[3] obtained a synthetic product of the same melting point as that of the natural lactone and, on this basis, suggested the structure (VI) for the lactone.



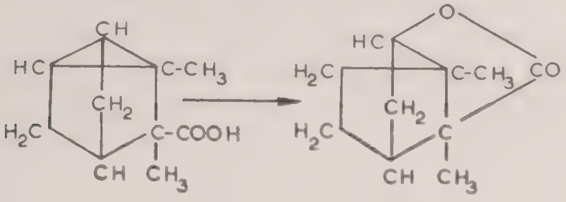
III
Nor-methyl camphor

IV

V

VI
Structure proposed by Bhattacharyya

Simonsen and Barton[4], however, proposed structure (IV). Their proposal is based on the observation that the lactone of tricycloekasantalic acid be obtained in exactly the same manner as the lactone (VIII) of a lower homologue, teresantalic acid (VII).

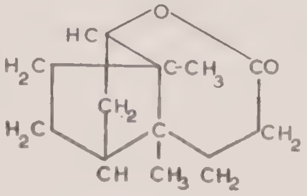


VII

Teresantalic acid

VIII

Lactone of teresantalic acid



IX

Structure proposed by Simonsen and Barton

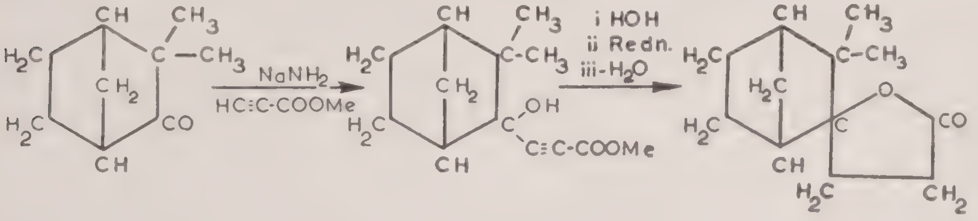
The lactone when subjected to alkaline hydrolysis yields an acid which, as revealed by permanganate oxidation, does not contain a secondary hydroxyl group. This fact makes structure IX untenable for the lactone.

The lactone, on reduction with lithium-aluminium-hydride, gives a diol, $C_{12}H_{22}O_2$ (b.p. 135-140°C./1.5 mm.), which on dehydration with acetic anhydride furnished an unsaturated alcohol, $C_{12}H_{20}O$ (b.p. 108-112°C./4.5 mm). It gives on treatment with hydrogen chloride in methyl alcoholic solution, a chloro-ester which on further treatment with dimethyl aniline yields an unsaturated ester (b.p. 93°C./1-2 mm ; $n^{26.5}$ 1.4810 ; $D_4^{26.5}$ 1.000 ; M_R 59.23 ; $[\alpha]_D$ $-4.12°$). On hydrolysis of this ester an acid (m.p. 105-106°C.) was obtained. This acid does not contain an exocyclic double bond as revealed by its infra-red spectrum. The reconversion of this acid into the lactone, on treatment with dilute sulphuric acid, disproves the validity of structure (VI).

The infra-red spectrum of the lactone measured in carbon disulphide showed maximum of 1775-¹cm. and suggested its being an γ-lactone of structure (XII).

Synthesis of the lactone of tricycloekasantalic acid

In order to establish the identity of the compound of the structure (XII) with the lactone of tricycloekasantalic acid, the compound was synthesised from camphenilone (X) according to the following scheme:



X

XI

XII

Camphenilone

Lactone of tricyclo-ekasantalic acid

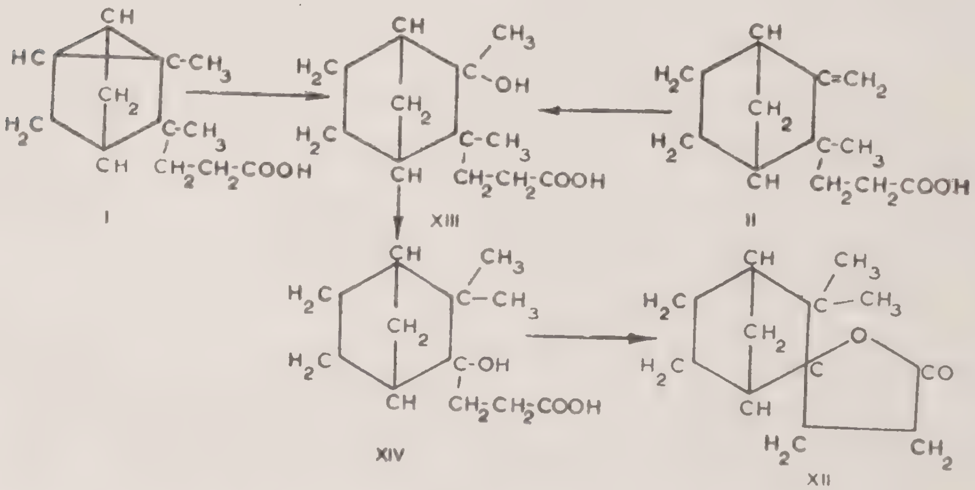
Camphenilone was reacted with methyl propiolate in presence of sodamide in liquid ammonia. The resultant product was hydrolysed with alcoholic potash, and the acid so obtained, was reduced with hydrogen using Adam's Catalyst. The reduced product consisted of a lactone and an acid. The acid was converted into the lactone by treatment with acetic anhydride. The total yield of the lactone (m.p. 102-103°C.) was 57 per cent of the unsaturated acid. Mixed melting point with the lactone (m.p. 102-103°C.) of tricycloekasantalic acid was the same. Its infra-red spectrum is also identical with that of natural lactone.

The synthetic lactone (XII) shows the reactions of the natural lactone. On treatment with hydrogen chloride in methyl alcoholic solution, a chloro-ester was obtained which underwent dehydro-chlorination with dimethyl aniline and furnished an unsaturated ester (b.p. 93°C./1-2 mm., $n^{22°}$ 1.4830 ; $D_1^{22°}$ 1.002 ; M_R 59.33 ; $[\alpha]_D$ $-9.38°$), which on hydrolysis, gave an unsaturated acid, (m.p. 105-106°C.) ; mixed melting point with the acid (m.p. 105-106°C.), similarly obtained from the natural lactone, remained undepressed.

These experiments establish the structure of the lactone of tricycloekasantalic acid as (XII). It is evident that during its formation from tricycloekasantalic acid (I), a structural rearrangement takes place.

Probable mechanism of the rearrangement

It would appear that the hydroxy acid (XIII) (or the carbonium ion derivable from it) is the intermediate during this rearrangement. This intermediate can be visualised as rearranging itself into (XIV) which can ultimately yield the lactone (XII). These considerations make it necessary to repeat Bhattacharyya's work, and to isolate the acid (XIII) and to see if the same lactone (XII) is obtained by treating the acid (XIII) with dilute sulphuric acid or concentrated formic acid. This is felt to be all the more desirable since Bhattacharyya's synthetic lactone has the same melting point as the natural lactone of tricycloekasantalic acid, and its identity has not been completely proved by mixed melting point and infra-red determinations. This work is now in progress.



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Changes in the Optical Rotatory Power of Camphor Derivatives

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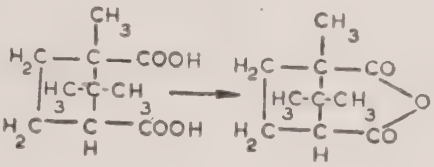
The study of the derivatives of aryl-iminocamphors has revealed the following relationships between the chemical constitution and optical rotatory power: (1) The formation or the opening of a ring produces a great change (a decrease or an increase) in the optical rotatory power. (2) Unsaturation generally leads to an increase in the rotatory power. This is remarkably illustrated in the case of the aryl-imino- and -bisimino derivatives of camphor, showing the effect of the optimum association of the azethenoid groups, conjugated linkings, and benzene rings within a given molecular compass on the optical rotatory power. The narrower the molecular compass containing a given number of conjugated double bonds, the greater the rotation. (3) A phenomenal and remarkable depression in the rotatory power occurs when the azethenoid group in the molecule of these compounds is reduced. For example, when *p*-iminocamphordiphenylamine is reduced to *p*-aminocamphordiphenylamine by the addition of two atoms of hydrogen, the molecular rotatory power, is phenomenally reduced from 8818° to 170°—a fall of 8648°.

The conditions which a molecule must satisfy, in order to exhibit optical activity, have been defined in their classical work by Pasteur, Van't Hoff, and Le Bel[1]. Besides molecules containing carbon atoms, those of about a score of other elements have been shown to exhibit optical activity under suitable conditions. Among the groups of natural products, the essential oils are characterised by containing optically active constituents. Camphor is one of these constituents: it has greatly aided and stimulated investigations in optical activity. It is one of the few substances which show optical activity in the gaseous, liquid and solid states.

Although a large amount of experimental work has been carried out on optically active compounds, except in a few directions, it is difficult to correlate this work as the mathematical physicist has not been able so far to give any workable theory of optical activity in relation to the chemical constitution of the molecule. The theories of optical activity put forward by Max Born, Kühn, Gray, de Mallemann, Boys, Condon and others give expressions for optical rotatory power containing several parameters which cannot be evaluated: they do not, therefore, enable us to calculate the value of optical rotatory power of chemical compounds which could be compared with those observed by direct measurement. "The real theory of optical rotatory power may be found by the mathematician, but is concealed from the chemist in the papers of Born", wrote Professor T. M. Lowry, "who recognized that *four* coupled electrons are required to produce optical rotatory power". The chemist has also not been able to draw any wide generalizations from this enormous amount of work. The property of optical activity is, thus, remarkably esoteric.

We will now discuss the optical activity of a few derivatives of camphor to illustrate the above mentioned points:

1. *d-Camphoric acid* (I) and *d-Camphoric anhydride*[2] (II)

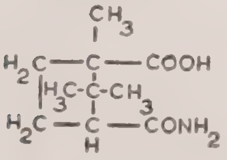


I II

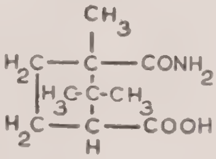
$[\alpha]^{35°c}_{5461} = +56.4°$ $[\alpha]^{35°}_{5461} = 0.0°$

The specific rotatory power of *d*-camphoric acid (I) is $[\alpha]^{35°}_{5461} = +56.4°$ in ethyl alcohol, whereas that of *d*-camphoric anhydride (II), derived from it, is almost nil (Lowry[3] gives $[\alpha]_D^{20°} = -1°$). It might appear that the camphoric anhydride has racemised. But this is disproved by the fact that on hydrolysis it yields the original acid of correct rotatory power and melting point.

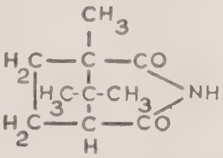
2. *d-Camphoramic acids* (III & IV) and *d-Camphorimide*[4] (V)



III



IV

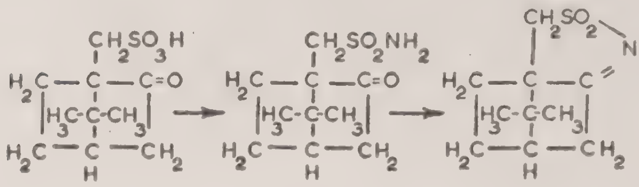


V

$[\alpha]_D^{20°} = +45°$ (α) $[\alpha]_D^{20°c} = +60.0°$ (β) $[\alpha]_D^{20°} = +6.1°$

d-Camphoramic acids exist in two isomeric forms α (III) and β (IV) owing to the unsymmetrical disposition of the two carboxyl groups in the molecule of camphoric acid. Whereas the specific rotatory power of the *d*-camphoramic acids is +45° (α) and +60.0° (β), that of *d*-camphorimide is only +6.1° in ethyl alcohol.

3. *d-Camphor-β-sulphonic acid* (VI), *d-camphor-β-sulphonamide* (VII) and *d-camphor-β-sulphon-anhydramide*[4] (VIII)

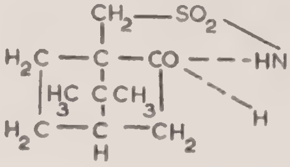


VI VII VIII

$[\alpha]_D^{20°} = +39.5°$ $[\alpha]_D^{20} = +1.5°$ $[\alpha]_D^{20°} = -33.5°$

The rotatory power of *d*-camphor-β-sulphonic acid (VI) in chloroform is +39.5°, that of the amide (VII) as low as +1.5° and the dehydrated product, namely, *d*-camphor-β-sulphon-anhydramide (VIII) −33.5° for which it has become laevorotatory.

It is thus seen that ring formation in the above mentioned compounds in this series leads to a phenomenal depression in rotatory power. In the case of derivatives of monosaccharides, the effect is opposite: the rotatory power of *d*-mannitol[5] is $[\alpha]_D{}^{20°} = -0.2°$, whereas that of *d*-mannolactone[6] is −53.8°. The high rotatory power of most of the sugars as compared with the corresponding polyhydric alcohols is a proof of the presence of the oxide ring in the former. Again the increase which is brought about in the rotatory power of sugar alcohols by boric acid is attributed to the formation of ring complexes. Other similar cases of both types may be cited. It may, therefore, be concluded that ring formation leads to a marked alteration in rotatory power which may be a decrease or an increase. The writer[8] formulated *d*-camphor-β-sulphonamide, in which the rotatory power is remarkably depressed as below (VIIa):



VIIa

It may be noted that the two hydrogen bonds (dotted lines) have been brought into action forming a loosely closed structure, which may account for its abnormally low rotatory power. In conformity with these ideas, the further conversion of *d*-camphor-β-sulphonamide (VII) into *d*-camphor-β-sulphon-anhydramide (VIII) with the formation of a ring, is again accompanied by great change in rotatory power (laevorotatory).

Configuration of the Molecule and Sign of Rotation

The Committee of the American Chemical Society on carbohydrate nomenclature adopted for dextro-rotatory glycerose the *d*-configuration and designated it *d*-glycerose and all compounds related to or derived from it were given the *d*-configuration. Thus ordinary grape sugar is *d*-glucose. It is dextrorotatory and has the *d*-configuration assigned to it, as it is related to *d*-glycerose.

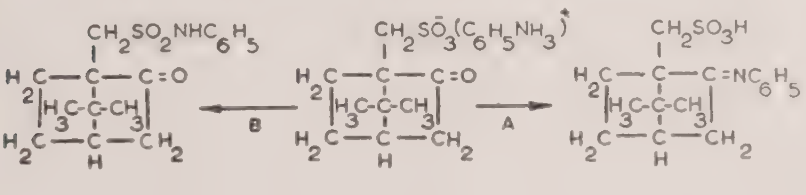
Fruit sugar, or fructose or laevulose is given the *d*-configuration as it is related to *d*-glucose, although it is laevorotatory. It is written as *d*-fructose. Its enantiomorph or mirror image is *l*-fructose, although it is dextro-rotatory. Among the compounds described in this paper there are *d*-camphor-β-sulphonanhydramide (VIII), 2-(N-phenyl ketimine)-*d*-camphane-10-sulphonic acid (XI), and *d*-camphorquinone (XIV) which have *d*-configuration because of their relation to *d*-camphor (dextro-rotatory), although they are laevorotatory.

A change in solvent is sometimes sufficient to bring about a change in the sign of rotation. Thus the specific rotatory power, $[\alpha]_D^{26°}$ of pure methyl-*d*-tartrate is $+2.1°$ but in benzene solution it is $-8.8°$. Again a 5 per cent solution of *d*-tartaric acid in different solvents gave the following values of rotatory power[9]:

Solvent	$[\alpha]_D^{20°}$
Water	$+14.4°$
Ethanol	$+\ 3.79$
Ethanol + Benzene (1:1)	$-\ 4.1$
Ethanol + Toluene (1:1)	$-\ 6.19$
Ethanol + Chlorobenzene (1:1)	$-\ 8.09$

It is, thus, clear that the sign of rotation of a compound is not related to its configuration. It must, therefore, be confessed that, at present, the magnitude and sign of rotation of a compound cannot be determined from its chemical structure and configuration. The relationship is much more complex. It is a puzzle and mystery, the solution of which is still an outstanding problem.

(4) *Anilino-d-camphor-β-sulphonate*[10] (IX), *d-camphor-β-sulphonyl-phenylamide*[11] (X) *and 2-(N-phenylketimine)-d-camphane-10-sulphonic acid*[12] *or anil* (XI)



X $[\alpha]_D^{35°} = +73.0°$

IX $[\alpha]_D^{35°} = +36.1°$

XI $[\alpha]_D^{25°} = -170.5°$

The specific rotatory power, $[\alpha]_D^{35°}$, of anilino-*d*-camphor-β-sulphonate in chloroform at 35° is $+36.1°$, *d*-camphor-β-sulphonyl phenyl amide $+73.0°$, whereas the corresponding ketimine or anil is strongly laevorotatory and has $[\alpha]_D^{25°} = -170.5°$. The anomalous mutarotation exhibited by the salts of *d*-camphor-β-sulphonic acid with organic primary nitrogen bases suggests that on dehydration they are capable of reacting in the ketimine form (reaction A) rather than in the sulphonamide form (reaction B), formulated above.

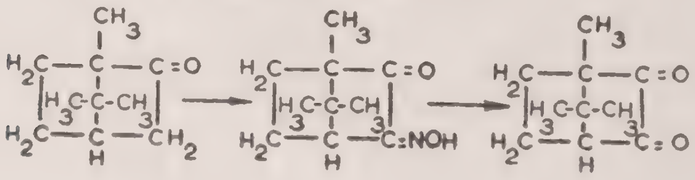
The present writer[13], in collaboration with Miss M. K. P. Amma, M.Sc., has studied the kinetics of mutarotation of the salts by following the reaction polarimetrically at constant temperature. They have used in this study, *o*-, *m*- and *p*-chloranilino-*d*-camphor-β-sulphonates, originally prepared by the writer and Shri O. N. Perti. The kinetics of the mutarotation were investigated at 25°C., 35°C. and 45°C. in chloroform in which solvent the mutarotation is most marked. In all cases the change involved in mutarotation is '*monomolecular*'. The ratio of velocity constants for 10° change in temperature was found to be about 1.4-1.5 in agreement with the view of Arrhenius[14] for ordinary reaction velocities.

It is also possible that the change may be due to amide formation (reaction B) or to both reactions A and B taking place simultaneously. This point was settled by studying the effect of temperature on the velocity constant (k) and applying the following equation of Arrhenius:

$$\frac{d \log k}{dT}=\frac{H}{RT^2}, \quad \text{Or} \quad \log k = C-\frac{A}{RT}$$

If the reaction were simple, namely, A or B, the plot of log k against 1/T (T in absolute degrees) would be a straight line according to the above mentioned equation. If, on the other hand, the observed reaction was a composite one, made up of two concurrent reactions A and B, the plot of log k against 1/T would not be a straight line. We found that in the case of each of these compounds a straight line was obtained indicating that the change involved in mutarotation was a simple one, A or B. The reaction, B, was excluded as the rotatory power decreased and in some cases even became laevo: the sulphonamides have greater rotatory power than the corresponding camphor-β-sulphonates without any change of sign of rotatory power (B), whereas the corresponding ketimines are laevorotatory (A).

5. *d-Camphor* (XII), *d-Isonitrosocamphor* (XIII), and *d-Camphorquinone*[2] (XIV)



XII XIII XIV

$[\alpha]^{35°}_{5461} = +°59.6$

stable (α) $[\alpha]^{35}_{5461} = +241.2°$

unstable (β) $[\alpha]^{35}_{5461} = +214.3°$

$[\alpha]^{35°}_{5461} = -165.5°$

The specific rotatory powers, $[\alpha]^{35}_{5461}$, in ethyl alcohol for the three compounds are recorded above. The *d*-camphorquinone which is configuratively related to *d*-camphor (dextro-rotatory) is strongly laevorotatory ; its enantiomorph, *l*-camphorquinone, is dextro-rotatory (+165.5°).

EFFECT OF CONJUGATION ON ROTATORY POWER[15]

(*a*) The marked effect of conjugated linkings on the optical rotatory power of derivatives of camphor, namely, the aryl-imino- and -bisiarylmino-camphors, is brought out very clearly by counting the number of conjugated double bonds of the six compounds shown in Table 1, and comparing their rotatory power in methyl alcohol. As the number of conjugated double bonds increases, the rotatory power increases. These examples strikingly illustrate the effect of the optimum association of azethenoid groups, conjugated linkings and benzene rings within a given molecular compass on rotatory power. The narrower the molecular compass containing a given number of conjugated linkings, the greater is the rotation.

(*b*) The phenomenal depression in the rotatory power which accompanies the reduction of the azethenoid group in the molecules of these compounds derived from *d*-camphor is illustrated in a most striking manner by the values of the rotatory power in pyridine recorded for the six compounds in Table 2.

TABLE 1—EFFECT OF CONJUGATION ON OPTICAL ROTATORY POWER OF CAMPHOR DERIVATIVES

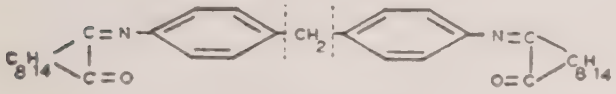
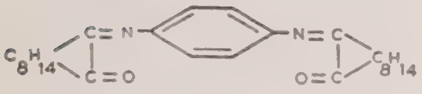
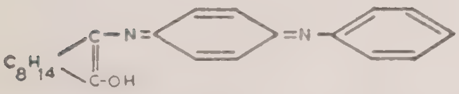
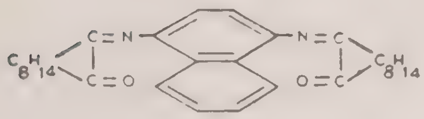
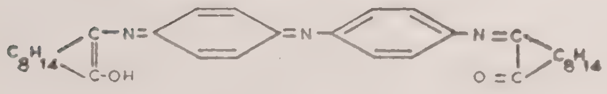
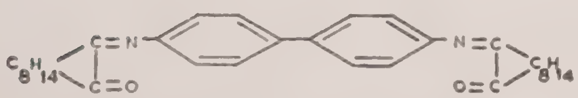
STRUCTURAL FORMULA	NUMBER OF CONJUGATED DOUBLE BONDS	$[M]_D^{35°}$	
		Dextro	Laevo
 a	2 × 5	+ 3990°	− 3941°
 b	7	5082	5078
 c	8	6303	6237
 d	9	9348	9356
 e	10	12460	12540
 f	10	5020	. .

TABLE 2—OPTICAL ROTATORY POWER OF CAMPHOR DERIVATIVES

Compound	$[M]^{35^\circ}$C. Hg Green	Difference
pp′-Diphenylmethanebisimino-*d*-camphor (I)	+6016°	5696°
pp′-Diphenylmethanebisamino-*d*-camphor	320°	
p-Phenylenebisimino-*d*-camphor (II)	8345°	8044°
p-Phenylenebisamino-*d*-camphor	301°	
p-Iminocamphor diphenylamine (III)	8818°	8648°
p-Aminocamphor diphenylamine	170°	

These phenomenal and remarkable changes in rotatory power effected by such slight alterations in molecular weight were unknown in the history of optical active compounds until camphor and its derivatives were brought into service in this field. The chemistry of some of the constituents of the essential oils and their derivatives has, thus, aided the study of stereochemistry in general and optical activity in particular.

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Estimation of Alcoholic Constituents and Esters in Essential Oils and Synthetic Mixtures —A Review

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The present position regarding the estimation of alcoholic constituents and esters in synthetic mixtures and natural essential oils has been reviewed. Attention has been drawn to the fact that for the estimation of alcohols in essential oils Cocking's formula should be used and not Gildemeister's formula, which is mathematically incorrect. The proposed formula for the estimation of two esters or two alcohols present in simple synthetic mixtures has been discussed.

Many essential oils contain esters and alcohols as the key constituents. One of the most widely used methods of estimation of an alcohol in an essential oil is the indirect method based on the estimation of the ester value of the oil before and after complete acetylation with an equal volume of pure acetic anhydride and fused sodium acetate. The practical details of the method have been fully described[1-3].

The following formula is used for the estimation of per cent free alcohol content (a) of an essential oil.

$$a=\frac{M(v_2-v_1)}{561.04-0.4201(v_2-v_1)} \qquad (1)$$

where M is the molecular weight of the main alcoholic constituent of the essential oil, and v_1 and v_2 are its ester values before and after acetylation. The error caused by applying formula (1) is well brought out in Nakao and Shibue's work on the oil of peppermint[2,4].

Cocking[5] appears to be the first to notice the discrepancy in the use of formula (1) originally due to Gildemeister. He deduced and suggested the use of formula (2).

$$\text{Percentage of free alcohol}=\frac{M(v_2-v_1)}{561.04-0.4201\,v_2} \qquad (2)$$

This formula, which is theoretically sound, seems to have remained unnoticed. It has not been mentioned in standard books on essential oils.

The procedures in vogue at present enable one to approximately estimate the total primary and secondary alcohols, or tertiary alcohols present in an essential oil in terms of a specific alcohol which is known to occur in the largest preparation in a particular essential oil.

In a previous communication[6], a critical study of Gildemeister's formula (1) and Cocking's formula (2) was made and their application to specific cases of mixtures of geraniol and geranyl acetate was carried out. It was shown, that for this system, the use of Gildemeister's formula results in a low alcohol per cent and that the per cent geraniol content of the mixtures may be off by as much as seven. On the other hand, Cocking's formula gave excellent results. Employing formylation technique of Glichitich[7], it was shown by Mathur *et al.*[8] that for a binary system consisting of benzyl alcohol-benzyl acetate, the percentage of benzyl alcohol calculated from Gildemeister's formula differs considerably from the actual value, thus proving that Gildemeister's formula is incorrect. The results obtained from Cocking's formula agree within one per cent of actual benzyl alcohol content of the mixture.

The extent of variations in the per cent of alcohol in essential oils and synthetic blends as determined by Gildemeister's formula and by the new formula due to Cocking are of importance from the commercial point of view. Jain and Lal[9] have calculated the extent of deviation of the observed percentage of an alcoholic constituent when calculated using Gildemeister's formula and when using Cocking's formula from the actual percentage present in the binary mixtures consisting of menthol-menthyl acetate and santalol-santalyl acetate.

They have also determined the deviation in the case of different samples of Indian vetiver oil and palmarosa oil based on actual determinations.

Benzyl alcohol has been estimated by the classical acetylation method in synthetic mixtures of benzyl alcohol and benzyl acetate by Pant *et al.*[10] Calculated from Cocking's formula (2) the per cent alcohol content was accurate within 1 per cent of the actual per cent benzyl alcohol content while the results obtained by the use of Gildemeister's formula showed considerable deviation ; the maximum deviation being as much as 8.2 per cent. Consider the case of a binary mixture consisting of *a* per cent of an alcohol A of molecular weight M_a and *b* per cent of ester B which is the acetate of alcohol A and hence has the molecular weight $(M_a+42.01)$.

For such binary mixtures, the maximum difference, 'y', between the observed per cent of an alcohol calculated from Gildemeister's formula and the actual per cent alcohol is given by the equation[10].

$$y=\frac{M_a d}{561.04-0.4201d}-a \qquad (3)$$

where $d=v_2-v_1$

and it can be shown that for maximum deviation

$$a=\frac{(M_a+42.01)\pm\sqrt{M_a(M_a+42.0)}}{0.4201} \qquad (4)$$

The maximum deviation between the observed per cent of alcohol calculated from Gildemeister's formula and the actual per cent alcohol present have been

TABLE 1—MAXIMUM DEVIATION OF VALUES OBTAINED BY GILDEMEISTER'S FORMULA FROM ACTUAL ALCOHOL PER CENT FOR SOME ALCOHOL-ACETATE SYSTEMS

SYSTEM	PER CENT ALCOHOL CORRESPONDING TC MAXIMUM DEVIATION	MAXIMUM DEVIATION
Geraniol-geranyl acetate	46.68	−5.92
Benzyl alcohol-benzyl acetate	54.14	−8.24
Santalol-santalyl acetate	45.47	−4.29
Phenyl ethyl alcohol-phenyl ethyl acetate	53.58	−7.36

calculated for some such binary systems from the above formula and the results are given in Table 1.

Formula and methods for the estimation of individual alcohols in a mixture containing more than one alcohol are not available in literature ; therefore work in this line was taken up.

Consider a mixture consisting of two alcohols A (mol. wt., M_a, per cent, *a*) and B (mol. wt., M_b, per cent, *b*) and ester C (mol. wt., M_c per cent, *c*). Let the ester value of pure acetate of alcohols A and B and ester C be V_a, V_b, and V_c respectively. Lal *et al.*[11] have on the basis of mass fraction concept derived the formulae (5) and (6) for the estimation of two individual alcohols in a mixture consisting of two monohydric alcohols and one ester.

$$a=\frac{M_a\left[v_1\left\{\frac{1}{v_c}-\frac{1}{v_b}+\frac{1}{1336}\left(1-\frac{v_2}{v_c}\right)\right\}+\left(\frac{v_2}{v_b}-1\right)\right]}{(561.04-0.4201v_2)\left[\frac{1}{v_b}-\frac{1}{v_a}\right]} \tag{5}$$

$$b=\frac{M_b\left[v_1\left\{\frac{1}{v_c}-\frac{1}{v_a}+\frac{1}{1336}\left(1-\frac{v_2}{v_c}\right)\right\}+\left(\frac{v_2}{v_c}-1\right)\right]}{(561.04-0.4201v_2)\left[\frac{1}{v_a}-\frac{1}{v_b}\right]} \tag{6}$$

In case a binary mixture consisting of two alcohols the formulae (5) and (6) reduce to the simple forms (7) and (8).

$$a=\frac{M_a\,(v_2-V_b)}{(561.04-0.4201\,v_2)}\cdot\frac{V_a}{V_a-V_b} \tag{7}$$

$$b=\frac{M_b\qquad v_2-V_a}{(561.04-0.4201\,v_2)}\cdot\frac{V_b}{V_b-V_a} \tag{8}$$

The set of formulae (7) and (8) have been verified[11,12] in the case of binary mixtures consisting of beta phenyl ethyl alcohol, cinnamyl alcohol and benzyl alcohol, cinnamyl alcohol and beta phenyl ethyl alcohol.

Recently the investigation[13,14] has been extended to more complex systems also. Consider a system consisting of a, b, c and k and d per cent of alcohols A, B, and ester C, ketone K and acid D respectively and let their respective molecular weights be M_a, M_b, M_c, M_k, and M_d and the ester value of pure acetate of alcohol A and B and ester C be V_a, V_b, and V_c respectively. Further let V_d be the acid value of pure acid D, then ;

$$a=\frac{M_a\left[v_1\left\{\frac{1}{V_c}-\frac{1}{V_b}+\frac{1}{1335.5}\left(1-\frac{v_2}{V_c}\right)\right\}+\left(\frac{v_2}{V_b}-1\right)+\frac{k}{100}\left(1-\frac{v_{2A}}{1335.5}+\frac{v_3}{V_d}\left(1-\frac{v_2}{1335.5}\right)\right)\right]}{(561.04-0.4201\ v_2)\ \frac{1}{V_b}-\frac{1}{V_a}} \tag{9}$$

and

$$b=\frac{M_b\left[v_1\left\{\left(\frac{1}{V_c}-\frac{1}{V_a}\right)+\frac{1}{1335.5}\left(1-\frac{v_2}{V_c}\right)\right\}+\left(\frac{v_2}{V_a}-1\right)+\frac{k}{100}\left(1-\frac{v_2}{1335.5}\right)+\frac{v_3}{V_d}\left(1-\frac{v_2}{1335.5}\right)\right]}{(561.04-0.4201\ v_2)\ \left[\frac{1}{V_a}-\frac{1}{V_b}\right]} \tag{10}$$

A survey of chemical literature did not reveal a method for estimation of two individual esters in a complex system. In a paper Lal *et al.*[15] have deduced the formula (11) and (12) for the estimation of two different esters in a system consisting of an alcohol A and two esters B and C and ketone K of respectively molecular weight M_a, M_b and M_c and M_k.

$$b=\frac{100v_1-(100-a-k)V_c}{V_b-V_c} \tag{11}$$

$$c=\frac{100v_1-(100-a-k)V_b}{V_c-V_b} \tag{12}$$

where V_a, V_b and V_c are the ester value of acetate of pure alcohol A, and pure ester B and C.

These formulae have been verified in the case of mixtures consisting of benzyl alcohol, benzyl acetate and santalyl acetate[15], and in the case of mixtures consisting of geraniol, benzyl acetate and beta phenyl ethyl acetate[16].

Experimental verification of these formulae on an extensive scale is in progress. During the course of the experiments practical methods of determination of alcoholic constituents in essential oils have also been studied. The formylation method of estimation was selected and a time-temperature study[8] of the course of formylation of different alcohols such as benzyl alcohol, geraniol, beta phenyl ethyl alcohol, menthol, dimethyl benzyl alcohol and linalool was made at a temperature of 3 to 5°C. Similarly, a study of the effect of time on the course of formylation of some primary, secondary and tertiary alcohols at 25°-30°C. (room temperature during summer) with ordinary formic-acetic anhydride reagent has been made[17].

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Antibacterial Activity of a Few Common Essential Oils

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The antibacterial activity of some essential oils has been examined against *S. aureus, E. coli, E. typhosa* and *B. dysenteriae*. It is suggested that they may prove therapeutically useful in the treatment of the infections of digestive tract in the form of their stable and non-irritant emulsions.

Essential oils on account of their high germicidal power, have found considerable application in disinfectant preparations and in surgery. Their antiseptic properties are due to their volatility and also partly to their solubility in lipoids which enable their ready penetration into the protoplasm. Earlier work by De and Subrahmanyan[1], De[2], and Subrahmanyan[3] has shown that essential oils like lemongrass oil and gingergrass oil have phenol coefficients of the order of 17, and 10, respectively, and that they would, consequently, be good disinfecting agents. The disinfectant action of a few essential oils from some native Australian plants, mainly eucalyptus, had been the subject of study by Penfold and Grant[4], but their results gave little indication of their activity. Atkinson and Reinsford[5], have reported on the antibacterial activity of essential oils of some myrtaceous plants. Apart from these no further information is on record regarding the antibacterial properties of these oils. Since oils, like lemongrass oil, possessing high phenol coefficient, have reputed use in medicine, a dozen of them were carefully tested by the well-known Oxford Cup Method[6], for their antibiotic activity against *Staphylococcus aureus, Escherichia coli, Eberthella typhosa* and *Shigella dysenteriae*.

EXPERIMENTAL

The samples of oils were freshly distilled and tests, such as specific gravity, refractive index, solubility in alcohol and in some cases optical rotation, were made in order to determine the conformity of the samples to specifications given in the British Pharmacopoeia[7]. The assay was performed according to the method described by Heatley[6]. The medium consisted of Lemco meat extract 1 per cent, bacteriological peptone 1 per cent, sodium chloride 0.5

TABLE I—ANTIBACTERIAL ACTIVITY OF ESSENTIAL OILS†

OIL	PHENOL COEFFICIENT *E. typhosa*	DIAM. OF THE ZONES OF INHIBITION			
		*S. aureus**	*E. coli*	*E. typhosa**	*S. dysenteriae**
Cedar wood	1.6	12,12	—	—	—
Cinnamon bark	7 to 9	14,13.9	17,17	—	14,14
Citronella	10	40,40.5	—	16,16	17,17.5
Clove	8	22.9,23	30,30.2	—	15,15.1
Eucalyptus	5	thin growth	30,29.9	24,24	sterile plate
Gingergrass	11.5	30,30	16,15.9	20,19.9	17,17.4
Lemongrass	18.5	sterile plate	17,17	18,18	15,15.5
Palmarosa	14	12,12	45,44.8	—	32,32
Patchouli	0.2	—	13,13	—	—
Pine	5.5	—	12,12	25,25.4	23,24
Sandalwood	1.5	—	12.8,13	—	—
Turpentine	0.8	25,25	40,39	half plate sterile	sterile plate

† Results of two tests are given for each oil. A dash indicates insignificant readings.
* National Collection of Type Culture.

per cent and agar 2 per cent, buffered at neutral *p*H with phosphate buffer. The assay measurements were made after incubation of the plates for sixteen hr. at 37°C. The results are given in Table 1.

It may be seen from the results that some essential oils like those of *citronella,* lemongrass, and gingergrass, have profound inhibitory activity on gram-positive organisms such as *Staphylococcus aureus* while oils from palmarosa, eucalyptus, and pine are highly inhibitory to the growth of gram-negative organisms like *Escherichia coli, Eberthella typhosa,* and *Bacillus dysenteriae*. Turpentine appears to inhibit the growth of both types of organisms quite contrary to what may be expected from its phenol coefficient value. In general, all the oils appear to possess antibacterial activity against the test organisms to a greater or less extent. The activity also appears to run somewhat parallel to the phenol coefficient of the oils, as is seen from the values given in column 2 of the Table. In the case of turpentine, however, the results are rather divergent. Since, in the case of different organisms, zones of inhibition with different diameters are obtained with the same oil under the same experimental conditions, it is clear, that diffusibility of the oil into the agar, which is usually a factor in the formation of zones of inhibition, has not affected these results.

Pronounced insolubility in water, the rather penetrating smell and, often, their intense irritant action, are among the serious impediments in the use of these oils in medicine. Nevertheless, it has to be recognized that the essential oils hold definite promise and possibilities provided the above defects are overcome. They also possess the additional advantage that they can be orally administered. Recent work[8] has shown that these oils can be processed into stable and non-irritating emulsions without lessening their antiseptic properties. As these preparations are comparatively easy to make and as they will be protein-free they hold out considerable possibilities of therapeutic application, especially in the treatment of the infections of the digestive tract.

SUMMARY

1. A few common essential oils have been assayed by the Oxford Cup Method for antibiotic activity and most of them have shown to possess high inhibitory activity against the test organisms *S. aureus, E. coli, E. typhosa, B. dysenteriae.*

2. The possibilities of therapeutic application which these oils seem to hold out on account of their antibacterial activity, coupled with the advantages which their emulsions have been shown to possess, have been pointed out.

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Bactericidal Property of Indian Essential Oils with Special Reference to Lemongrass Oil

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Several Indian essential oils were tested for their bactericidal efficiency by the R. W. Test and by Garrod's modified method of Chick-Martin test in presence of different kinds of organic matter. Lemongrass oil was found to be the most active. Notable reduction in germicidal activity was, however, observed in the presence of organic matter.

The bactericidal efficiency of the essential oils was measured against different organisms and was found to be generally high against Gram-negative bacteria but very low against Gram-positive organisms. The activity against the Acid-fast organisms was practically nil.

The influence of several important factors on the germicidal efficiency of lemongrass oil emulsions was studied. It was demonstrated that all other factors being identical, the bactericidal efficiency of lemongrass oil was directly proportional to the citral content. The emulsifiable property of the oil and the efficiency of the emulsifying agent used were the other two important factors affecting its bactericidal action. Triethanolamine or potassium oleate and rosin soap were found to be more efficient than the other emulsifiers examined. Particle size and germicidal potency of the emulsions were correlated. No direct relationship was found between the viscosity of the emulsion and the bactericidal action. The *p*H of emulsified disinfectants was important from the point of view of the stability of emulsion. Use of an emulsifying agent in concentrations higher than the optimum was found to lower the bactericidal action of lemongrass oil emulsions. The solvent used in the preparation of disinfectant solutions of the oil, which on dilution form emulsions, was found to lower their bactericidal action as also the stability of the emulsions formed.

The important constituents of Indian essential oils were isolated and examined for R.W. coefficients. The relationship between the bactericidal action and the chemical constitution of the compounds was studied. The -CHO group was found to be more active than the -OH group. The presence of an α-β double bond in terpene aldehydes and alcohols was found to enhance the efficiency of the active group.

Lemongrass oil was found to be susceptible to autoxidation and the loss of citral and germicidal property during storage. It was demonstrated that the fall of citral and bactericidal potency was principally due to the autoxidation of the oil. The disinfectant preparations from lemongrass oil were found to lose their bactericidal efficiency on keeping. Addition of certain antioxidants such as ethyl or *n*-propyl gallate in conjunction with citric or tartaric acid, to the pure oil was found to retard the oxidative deterioration of the oil.

The work was carried out at the Department of Biochemistry, Indian Institute of Science, Bangalore, under the auspices of the Council of Scientific & Industrial Research.

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New Processes for the Production of Aromatic Chemicals Directly from Coal

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The coal carbonization industry—the only present-day source of aromatic chemicals—will not be in a position to meet the ever-expanding demand from various industries. Petroleum industry may only give a temporary relief. The processes which will supply the future demand for aromatic chemicals are the processes of hydrogenation and oxidation of coal. Both the processes have passed the laboratory stage ; the former has reached the semi-commercial scale while the latter is in the pilot plant stage. Hydrogenation of coal will produce large quantity of common aromatic chemicals, in addition to large quantity of polynuclear and heterocyclic compounds. Aromatic acids, mainly polycarboxylic acids of benzene, could now be produced by the direct oxidation of coal.

The demand for aromatic chemicals is increasing every day. Besides the essential oils industry, industries like dyes, resins, plastics, pharmaceuticals, insecticides and explosives also directly or indirectly, largely depend on these chemicals. The only major source of these products at present is the coal gas fraction obtained in the coal carbonization industry. But as the attention of this industry is mainly devoted to preparation of good quality coke, the chemicals are always obtained as by-products. It has been felt that alternative independent processes of production should be developed in order to meet their growing demand.

Hydroforming of petroleum

A process known as 'Hydroforming', using petroleum as raw material, was developed during the second world war for the production of toluene. The process involves the formation of a ring structure from the straight chain hydrocarbons, followed by dehydrogenation. Hydrocarbons including benzene, xylene, ethyl benzene, propyl benzene and cumene are obtained as by-products. Although petroleum appears to be a promising alternative source of aromatic chemicals, it is not likely to be exploited for their long term production mainly because the chemicals are produced at the cost of fuel and the oil reserves of the world are limited. On the other hand, deposits of coal are extensive and development of atomic energy will reduce its utilization as fuel.

It is considered that coal is a highly polymerised natural product having an elaborate network of carbon rings[1]. Methods of hydrogenation[2] and oxidation[3] have been found to be the best means of splitting carbon rings from the complex structure of coal. Both the processes have passed the laboratory stage ; the former has gone to semi-commercial scale while the latter is at the pilot plant stage.

Hydrogenation of coal

The coal hydrogenation process, first invented by F. Bergius, was developed for the production of synthetic petrol[4] and reached its peak during the second World War[5]. It is considered that hydrogenation of coal will not be economical for the manufacture of synthetic petrol but it can be used for the production of more costly aromatic chemicals. It produces chemicals like benzene, toluene, xylenes, naphthalene, phenol, cresols and xylenols in addition to polynuclear and hetero-cyclic compounds. The yields obtained are much more than those obtained in coking process.

The Carbide and Carbon Chemical Company of America, who have been perfecting the process for nearly 20 years announced their project in 1952[6-9]. The new process differs from the old method in the degree of hydrogenation. The hydrogenation of coal in the new process is completed at the liquid phase stage. It is considered that the nature of the catalysts used and other details of the process also are different[9]. It appears that the separation of solids from liquids, due to their fine dispersion[10], is presenting difficult problems which are yet to be solved. The liquid products obtained are divided into the following fractions: hydrocarbon, a phenolic and a nitrogenous fraction. The nitrogenous fraction has special significance because of the unusual stability of nitrogen in coal[11]. So far, 129 chemicals have been identified in the liquid, of which 32 are aromatic hydrocarbons, 27 aliphatic hydrocarbons, 24 phenols and 46 nitrogenous compounds[12].

Economic aspects

It has been estimated that compared to the coking process this process will produce from a given quantity of coal about 300-500 times more quinoline, 100-200 times more cresols, 60-80 times more phenol, 3-8 times more naphthalene and large quantities of aniline which is not recovered from the carbonization industry. Of the various compounds such as indane, acenaphthalene, fluorene, anthracene, phenanthrene, indole-4, indole-5, 3-phenylphenol, 2-methylquinoline, carbazole, 2-methyl-6-ethylcarbazole, 3-methylindole and di-benzofurane identified, some are present in economically recoverable quantities. At present some of the chemicals are produced using costlier synthetic methods.

The gaseous hydrocarbons from C_1 to C_{11} can be used for the production of aliphatic compounds. Sulphur can be recovered from hydrogen sulphide and ammonia can be converted into ammonium sulphate. The coke obtained can be used for making carbon electrodes.

Oxidation of coal

For the production of chemicals, the oxidation reaction is carried out at low temperature in different stages. At the initial stage oxygen is added resulting in the formation of oxygen containing groups. In the second stage considerable portion of coal is converted into products known as humic acids. In the third stage these acids are transformed into water soluble acids. This is the most important stage for the production of aromatic acids. The Coal Research Laboratory of Carnegie Institute of Technology, U.S.A., has installed a pilot plant for the production of aromatic acids by direct oxidation of coal. The main product of the process[13-16] is a mixture of aromatic acids of the benzene carboxylic series.

The acids can be used for the production of glyptal type of resins, and their esters can be used as plasticizers and lubricants.

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GERANIUM OIL

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India embraces vast tracts of tropical plains, temperate hills and valleys, with extremely suitable soil conditions and an ideal combination of moist and dry climate. There is unlimited scope for growing a variety of essential oil plants. The essential oil plantation on the Shevaroy Hills, which are particularly suitable for such plantations, was started in 1945 and experiments on raising geranium, patchouli, vetiver, davana, rosemary, peppermint, palmarosa, *Eucalyptus globulus* and *E. citriodora* were carried out. The experiences in connection with the cultivation and extraction of geranium are recorded here.

Geraniaceae species of *Pelargonium* is a native of the dry rocky slopes of South Africa. It is cultivated particularly in Algeria, South of France, Italy, Bourbon, and in Russia on the Caucasian coast of the Black Sea.

In Yercaud on the Shevaroy Hills, near Salem (South India) it is cultivated in open fields at an altitude of 4,500 ft. and above. Here mostly *Pelargonium capitatum* (Linn) Ait., and *Pelargonium graveolens* L'Her., to a certain extent, are cultivated. Geranium is propagated by cuttings in nursery baskets or beds and transplanted in newly cleared fields in the months of July and August.

The plant takes six to eight months to grow into a fine green bush. About seven months after planting, in the months of February and March, the first harvest is made. The green part of the plant is pruned carefully and distilled while still fresh. It yields an oil of great commercial value, widely used in soap and toilet industry. The yield varies from 0.143 to 0.21 per cent depending upon the abundance of leaves, as the major portion of the oil is contained in the leaves. The distillation is carried out over a period of six hours. Water-steam distillation process is employed in directly heated stills.

The oil is light yellowish green in colour and dissolves in 3 volumes of 70 per cent alcohol. It has sp. gr.$^{29°}$, 0.894 ; $n^{29°}$, 1.4672 ; acid val., 5.50 ; ester val., 54.75 ; and ester val. after acetylation, 205.70.

Green leafy shoots suitable for distillation are harvested thrice every year, in February-March, June-July, and October-November, which are generally the distillation seasons. It is also possible to so regulate harvesting that a continuous supply of material is maintained for distillation throughout the year if a large area is brought under cultivation with favourable weather conditions and well-spread rain. Normally plants are ready for harvesting after every four months. On the basis of acreage a well-maintained plantation may be depended upon to give a yield of 15 to 20 lb. of oil per acre from three collections. In one of the plots established on a newly opened and well-manured area in our estate the yield was as high as 30 lb. per acre.

The geranium plant stands in the fields for three to five years. The yield of green leaf tends to decrease after 3 years. By adopting proper methods of cultivation with chemical manure and compost, however, the life of the plant may be prolonged up to seven years. In Algeria the plant lives for twelve years.

In India also the span can be considerably lengthened if periodical tests of soil are carried out to study the actual needs of the geranium plant, and adequate measures for manuring are undertaken.

The geranium plant is sometimes a victim of fungi: *Pythium* sp., *Rhizoctonia solani* and *Fusarium* sp. Due to excess of rains, the leaves, stumps and also roots are affected. The spread of the disease can be controlled by pruning the plant at the early stage of the attack. If, however, the root is attacked, the plant dies. The entire plant must, therefore, be rooted out and burnt. Spraying with ½ per cent Bordeaux mixture or other fungicidal mixtures in a very dilute form is also effective.

At present an area of 200 acres is under geranium cultivation in the Shevaroy Hills. The yield of oil from this area is far below the total requirements of the country which are met by large imports every year. It is estimated that the Indian soap and perfume manufacturers require about 20,000 lb. of geranium oil annually. It is possible to produce the entire quantity required in the country with the help of the Central and the State Governments. Government can encourage the essential oil plantations by conducting soil research and offering such technical advice as may prove advantageous from time to time. The Shevaroy Hills appear to possess unlimited potentialities for large scale production of geranium oil, besides a variety of other essential oils, and may be exploited for the development of the essential oils industry.

GRASS OIL

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Many varieties of essential oil yielding grasses grow in large quantities on hill sides, valleys and in the plains of India, but only a negligible portion of these grasses is being utilized for distillation and extraction of oil. If an organization is set up to collect these useful grasses and distil essential oils from them, we will be in a position not only to meet the entire requirements of our soap and perfumery industries, but also to export an appreciable quantity.

In Courtallam (Tirunelveli District) its surroundings and all over Travancore State, *Cymbopogon caesius* Stapf. grows in abundance over many thousands of acres. The grass yields a very good essential oil of an agreeable odour, suitable, particularly, in the manufacture of washing soaps on commercial scale. Due to its known usefulness in skin diseases it may also be incorporated in bath soaps. If well-organized distilleries are set up in suitable places, uniform quality of the oil on a large scale can be prepared and may be used in place of the Ceylon citronella oil.

The oil (yield, 0.7 per cent) is pale yellow in colour, possesses an agreeable aroma and is soluble in an equal volume of 90 per cent and 8 to 9 volumes of 80 per cent alcohol. It has: sp. gr.$^{30°}$, 0.9221 ; $n^{30°}$ 1.4880 ; $[\alpha]_D$, 35°6′ ; acid val., 1.0 ; ester val., 23.4 ; ester val. after acetylation, 113.8 ; free alcohol content calculated as geraniol, 26.9 and total alcohols calculated as geraniol, 33.41.

The development of this industry will provide employment to a good number of people. Soap manufacturers in India may help the industry by using only indigenous essential oils, and the Essential Oils Research Committee of the Council of Scientific & Industrial Research may consider the question of necessary Government help to this industry during the second Five-Year Plan period.

β-IONONE FROM CITRAL

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The basic starting material for the commercial synthesis of vitamin A is β-ionone, which is converted into a C_{14}-aldehyde. The latter is then converted, through a series of steps, into vitamin A.

The first instance of ionones occurring in nature was given by Naves[1] who found the presence of β-ionone, α-ionone, cis-dihydroionone and isomeric ketones in costus (*Saussurea lappa*) roots. The economic production of β-ionone, however, has been rendered possible by the large availability of lemongrass oil. By treating lemongrass oil with aqueous sodium sulphite in an alkaline medium, a fairly good yield of citral has been obtained. The citral when treated with acetone and baryta and worked, gave pseudo-ionone. A study has been made on the cyclisation of pseudo-ionone by various acidic reagents to obtain mixtures of α- and β-ionones. Particular attention has been given to the influence of the cyclising agent on the preferential yield of β-ionone and amongst the acidic reagents used, concentrated sulphuric acid has been found to be the most suitable for obtaining a high yield. The economic aspect of the problem has also been surveyed.

REFERENCE

1. NAVES, *Helv. chim. acta,* **32** (1949), 1064.

PREPARATION OF AMYL CINNAMIC ALDEHYDE

NIRMAL SEN & S. SENGUPTA
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Amyl cinnamic aldehyde, obtained from castor oil, is an important constituent of jasmine perfume, floral bouquet and soaps. On pyrolytic treatment of castor oil, undecenoic acid and heptaldehyde were obtained in a yield of 26 and 30 per cent by volume respectively. The heptaldehyde on condensation with toluene gave amyl cinnamic aldehyde in a theoretical yield of 65 per cent. Further investigations are being carried out with a view to increase the yield of these products.

Castor oil and toluene are indigenous raw materials. The bulk of the castor oil exported to western countries is used for the preparation of costly aromatic chemicals like undecalactone, pentadecanone, γ-n-heptylbutyrolactone, γ-n-amylbutyrolactone, methylheptine carbonate and amyl cinnamic aldehyde.

A SPRAY REAGENT FOR THE DETECTION OF ESSENTIAL OIL CONSTITUENTS

G. S. MATHUR, M. S. BHATNAGAR & D. R. DHINGRA
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Alkaline potassium permanganate has been found to be a suitable spray reagent for the chromatographic detection of essential oil constituents. It gives spots with geraniol, nerol, linalool, *d*-limonene, geranyl acetate, benzyl acetate, citral and other aldehydes of C_{10}, C_{11} and C_{12} series.

The method consists in spotting the essential oil constituents in neutral alcohol with a fine capillary tube on Whatman filter paper No. 42 and spraying it with a solution of 1 per cent potassium permanganate in 2 per cent sodium carbonate (50 : 50) in water. Bright yellow spots are produced in the pinkish background.

ADULTERATION OF PHARMACEUTICAL PREPARATIONS OF ESSENTIAL OILS IN UTTAR PRADESH

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Examination of a large number of samples of pharmaceutical preparations containing essential oils received for analysis during the period 1950-55 indicated that in the case of single substance preparations the majority of the samples found unacceptable contained smaller amounts of active constituents than prescribed by the specifications ; only a few contained turpentine oil as an adulterant. In the case of mixtures of essential oils, ethyl alcohol was also found as an adulterant.

SEPARATION OF GERANIOL FROM PALMAROSA OIL UNDER LOW PRESSURE

C. KHUDADAD
Purandad Essential Oil Plantation, Doiwala, Uttar Pradesh

Palmarosa oil was distilled under reduced pressure with a view to produce a product very rich in geraniol. The distillate obtained, though answering the requirement of chemically pure oil, had poor aroma and did not come up to the specifications of the perfumers. This is probably due to the fact that a small quantity of esters, responsible for the pleasant odour and retained during steam distillation, was separated under vacuum distillation.

SYNTHETIC STUDIES IN DITERPENOID RESIN ACIDS

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One isomer of C_{12}-acid prepared synthetically has been found to have identical melting point and mixed melting point with the C_{12}-acid isolated from abietic acid. But infra-red studies indicate that it is different from the latter. Attempts have been made to synthesise the two isomers. Starting from Stork's ketone (1), the compound which should correspond to abietic acid in stereochemical configuration has been prepared.

Starting from the unsaturated ester (III) a crystalline product (IV) has been prepared whose structure has been established from analysis, infra-red studies and other properties. This on dehydration gives the unsaturated compound (V) which is being oxidised with the expectation of obtaining (II) in the other isomeric form corresponding to podocarpic acid.

SANTALOL AND ITS DERIVATIVES

B. B. GHATGEY, K. K. CHAKRAVARTI & S. C. BHATTACHARYYA
National Chemical Laboratory, Poona

Teresantalic acid has been converted into tricycloekasantalic acid by a new route in three unambiguous stages. A new method of synthesis of santalol from this acid is nearing completion.

Indian Vetiver Oil Industry. D. R. DHINGRA, G. N. GUPTA & GANESH CHANDRA (Kanpur).

Indian Soap J., **18** (1952), 77.—Vetiver oil is obtained by distillation of the root of *Vetiveria zizanioides* Linn. (*khus*). The plant occurs wild in North India and the oils prepared at different centres vary in their physico-chemical properties. In South India, vetiver is cultivated and the oil distilled from it resembles the oils from Java and Bourbon.

Analyses of a large number of samples of vetiver oils collected from various important centres show that the properties vary widely. The optical rotation of all the North Indian samples is laevorotatory, while South Indian and Java oils show dextro-rotation. (*Abstract*)

Indian Jasmine Oils. D. R. DHINGRA, G. N. GUPTA & U. N. SHUKLA (Kanpur).

1. *Indian Soap J.*, **16** (1951) 235 ; 259 ; 2. *Perfum. essent. Oil Rec.*, **44** (1953), 11 ; 3. *Perfum. essent. Oil Rec.* **42** (1951), 369.—There are three main varieties of jasmine, *Jasminum grandiflorum* (*chameli*). *J. sambac* (*bela* or *mogra*) and *J. auriculatum* (*juhi*), whose flowers are abundantly available in Uttar Pradesh. The flowers are mainly used for making hair oils and *attars*. The oils obtained from the flowers of *chameli*[1], *bela*[2] and *juhi*[3] have been examined. The analytical data have been recorded. (*Abstract*)

Oil of Champaca. D. R. DHINGRA, G. N. GUPTA & U. N. SHUKLA (Kanpur).

Ätherischeöle Riechstoffe Parfumerien Essenzen & Aromen, May 1953, 99. The oil of champaca is obtained from *Michelia champaca* Linn. (*champa*) flowers. The tree grows throughout south eastern tropical Asia, particularly in Java, the Philippines and India. The flowers are golden yellow in colour and emit a very pleasant odour. The oil (yield, 0.064-0.68 per cent) prepared by distillation of the flowers had the following characteristics: sp. gr.$^{30°}$, 0.9614-0.963 ; $n^{30°}$, 1.4925-1.4959 ; acid val., 4.17-6.22 ; sap. val., 69.02-77.23, and sap. val. after acetylation, 120.8-129.8. The oil was found to contain cineole, isoeugenol, benzaldehyde, phenylethyl alcohol and methyl anthranilate. (*Abstract*)

Essential Oil from *Acorus calamus* Rhizomes. J. C. GUPTA, G. N. GUPTA & D. R. DHINGRA (Kanpur).

J. Oil Tech. Ass. India, **11** (1956), 31.—*Acorus calamus* Linn. is indigenous to India and Burma. The oil from the dried rhizomes possesses good germicidal properties and is also employed in perfumery. The physico-chemical constituents of the oil have been examined. The oil consists mainly of asarone, 80.1 per cent along with calamene, eugenol methyl ether, calamenenol and palmitic and other fatty acids. (*Abstract*)

Essential Oil of *Ocimum basilicum*. S. N. DHINGRA, D. R. DHINGRA & G. N. GUPTA (Kanpur).

Indian Soap J., **19** (1954), 251.—Essential oils from the leaves and flowers of sweet basil (*Ocimum basilicum*) grown under direct supervision have been extracted and examined. The oil from the leaves (yield, 0.16-0.34 per cent) contains methyl cinnamate, 43.6-63.6 ; alcohols as linalool, 12.0-33.8 ; methyl chavicol, 10.2-23.3 and ocimene, 0.8-4.5 per cent. The oil from the flowers (yield, 0.3-0.54 per cent) is composed of methyl cinnamate, 47.7-74.3 ; alcohols as linalool, 8.6-28.5 ; methyl chavicol, 6.5-21.9 ; and ocimene, 5.5 per cent. (*Abstract*)

Essential Oil of Kewda. S. N. DHINGRA, U. N. SHUKLA, G. N. GUPTA & D. R. DHINGRA (Kanpur).

Perfum. essent. Oil Rec., **42** (1951), 114 ; **45** (1954), 219.—*Kewda* (*Pandanus odoratissimus*) flowers grow abundantly in South Orissa and Uttar Pradesh, and are extracted for their fragrant oil at both the places. It has been observed that the yield of oil from U.P. flowers is about half of that obtained in South Orissa where it varies between 0.025 to 0.030 per cent on the weight of fresh material. The oils prepared by steam distillation have been examined and their properties and constituents have been recorded. (*Abstract*)

Otto of Rose. G. N. GUPTA, GANESH CHANDRA & D. R. DHINGRA (Kanpur).

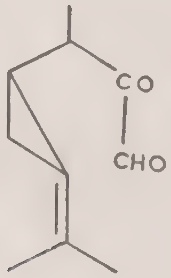
Indian Soap J., **21** (1955), 61.—Two varieties of rose, *Rosa damascena* and Rose Edward, are grown in North India. The former is grown mainly in Aligarh and Ghazipur, while the latter is raised at Kanauj and other places. The yield of oil on distillation of both the varieties was found to be 0.02-0.04 and 0.01-0.016 per cent respectively. The oils were also prepared by extracting the flowers with benzene and steam distilling the concretes obtained. The results of the examination of the oils have been recorded. (*Abstract*)

Chemical Examination of Indian Spearmint Oil : Part I—Separation and Characterization of Various Constituents. K. K. CHAKRAVARTI & S. C. BHATTACHARYYA (Poona).

Perfum. essent. Oil Rec., **45** (1954), 217.—Indian spearmint oil, isolated from *Mentha viridis* Linn. has been chemically investigated for the first time. The plant, grown under strict botanical control, on steam distillation gave 0.23 to 0.31 per cent of a pale yellow oil. Maximum yield was obtained by using mixed fertilizers and amounted to 60 lb. per acre. The oil has been found to contain traces of α-pinene, phellandrene, dihydrocarvol and its acetate and 31-34.6 per cent of 1:8 cineole. Its main constituent (47.6-54.4 per cent), however, is a new monoterpenic glyoxal ($C_{10}H_{14}O_2$). This new monoterpenic derivative has been named mintglyoxal ; it forms only monosemicarbazone and shows strong absorption in the ultra-violet region. It is probably the first specimen of a glyoxal derivative isolated from essential oils. Indian spearmint oil also contains a non-semicarbazone forming liquid (8-9 per cent), which is somewhat similar to mintglyoxal and shows strong absorption in the ultra-violet region. (*Abstract*)

Chemical Examination of Indian Spearmint Oil : Part II—Elucidation of the Structure of Mintglyoxal. K. K. CHAKRAVARTI & S. S. BHATTACHARYYA (Poona).

Perfum. essent. Oil Rec., **46** (1955), 256.—From an extensive chemical and physico-chemical study of mintglyoxal, its derivatives and degradation products, it has been found to possess the following structure:



Ultra-violet and infra-red spectra of this compound and its derivatives were very helpful in this investigation.

Mintglyoxal is the first example of a terpenic material containing an unattached cyclopropane ring. (*Abstract*)

Chemical Examination of Malabar Lemongrass Oil : Part I—Characterization of Lower Boiling Fractions. K. K. CHAKRAVARTI & S. C. BHATTACHARYYA (Poona).

Perfum. essent. Oil Rec., **45** (1954), 184.—The essential oil from *Cymbopogon nardus* Rendle, locally known as 'Malabar lemongrass oil', has been critically examined. The lower boiling fraction contains *l*-α-thujene, *l*-α-pinene, *l*-camphene, *l*-limonene, *l*-borneol and *l*-α-terpeneol. Camphene and borneol together form nearly 70 per cent of the lower boiling fraction or 36 per cent of the total oil. *l*-α-Thujene, which is present in this oil, does not appear to have been isolated before from natural sources. (*Abstract*)

Chemical Examination of Malabar Lemongrass Oil : Part II—Characterization of Higher Boiling Fractions. N. B. GHATGEY, R. K. RAZDAN, K. K. CHAKRAVARTI & S. C. BHATTACHARYYA (Poona).

Perfum. essent. Oil Rec., **46** (1955), 365.—The higher boiling fraction of Malabar lemongrass oil has been separated into seven distinct constituents: Hydrocarbon A, hydrocarbon B, alcohol A, oxygen containing compound B, higher boiling fraction A, higher boiling fraction B and higher boiling fraction C.

The hydrocarbon A ($C_{15}H_{24}$) is predominantly a non-cadinenic tricyclic sesquiterpene with one double bond.

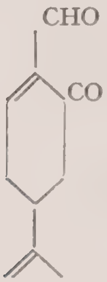
The hydrocarbon B ($C_{15}H_{24}$) is a cadinenic hydrocarbon with two double bonds, one of which is in the form of a methylene group. Oxygenated compound B is not an isoprenoid. From degradative and other experiments it has been characterized as elemicin. (*Abstract*)

Chemical Examination of Indian Dill Oil—Isolation of Dihydrocarvone. K. K. CHAKRAVARTI & S. C. BHATTACHARYYA (Poona).

Indian Pharm., **9** (1954), 218.—Indian dill (*Anethum sowa* D.C.) *seed oil* has been found to contain *d*-limonene, 34.4 ; *d*-carvone, 30 and dill apiole, 18.9 per cent. A new constituent, *l*-dihydrocarvone, has been isolated from the oil for the first time. It occurs in the oil to the extent of 9 per cent. The apiole-free lower boiling fraction of the oil with higher carvone content (37 per cent) may be accepted in the pharmacopoeia. (*Abstract*)

Oxidation of Carvone and Citronellal with Selenium Dioxide. K. K. Chakravarti & S. C. Bhattacharyya (Poona).

Perfum. essent. Oil Rec., **46** (1955) 341.—In connection with the investigations on the preparation of compounds analogous to mintglyoxal obtained from Indian spearmint oil, oxidation of carvone and citronellal with selenium dioxide has been carried out. Carvone produces two compounds: (I) and (II) along with a highly oxygenated material, $C_7H_{6-8}O_4$, which could not be characterized.



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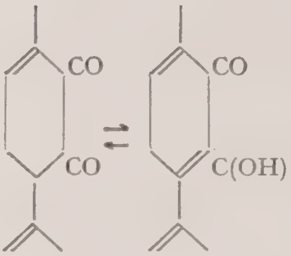

II

Citronellal on similar treatment produces an α-β-unsaturated dialdehyde and not the glyoxal as expected.

All the compounds and their derivatives have been thoroughly investigated. (*Abstract*)

Longifolene Fraction from Indian Turpentine Oil. B. B. Ghatgey & S. C. Bhattacharyya (Poona).

Perfum. essent. Oil Rec., **47** (1956), 122.—Several derivatives of longifolene have been prepared. Two of these are lactonic in nature and likely to throw some light on the structure of this hydrocarbon.

Another new hydrocarbon containing two double bonds has also been isolated from the longifolene fraction. (*Abstract*)

Cinnamyl Alcohol and its Derivatives. K. K. Chakravarti & S. C. Bhattacharyya (Poona).

J. Indian chem. Soc., industr. Edn, **13** (1955), 31.—The method of preparation of cinnamyl alcohol from cinnamic aldehyde using aluminium alkoxide as the reducing agent has been standardized. Yield of alcohol is about 80 per cent. Several improvements as regards experimental technique and quantity of solvent to be used have been incorporated. (*Abstract*)

Musk and Allied Compounds. K. K. CHAKRAVARTI & S. C. BHATTACHARYYA (Poona).

Indian Patent No. 58,203.—A short route for the synthesis of musk acid starting from easily available raw materials, has been standardized. Musk acid is a suitable material for the synthesis of muscone. (*Abstract*)

Synthetic Approaches Towards Civetone. V. K. BALAKRISHNAN, U. G. NAYAK, R. K. RAZDAN, K. K. CHAKRAVARTI & S. C. BHATTACHARYYA (Poona).

Indian Patents 55,504, 55,757, 56,391 and 57,541.—A short route for the synthesis of civetone dicarboxylic acid has been standardized. It is a suitable material for the synthesis of civetone. (*Abstract*)

Infra-red Spectra of Compounds Containing Cyclopropane Ring & its Application to Quantitative Evaluation of Sandalwood Oil. (MRS.) S. DASGUPTA, V. K. BALAKRISHNAN, K. K. CHAKRAVARTI & S. C. BHATTACHARYYA (Poona).

Perfum. essent. Oil Res., **46** (1955), 330.—Infra-red spectra of α-santalol, tricycloekasantalic acid, teresantalic acid, tricyclene and Δ^3-carene have been investigated. These, along with mintglyoxal and several of its well-characterized derivatives, form a representative group of compounds containing cyclopropane ring.

Mintglyoxal and its derivatives contain an unattached cyclopropane ring. In Δ^3-carene, the cyclopropane ring is fused to a cyclohexane nucleus. It also contains a gem-dimethyl group. In α-santalol and other related compounds examined cyclopropane ring forms part of a highly complicated tricyclic ring system.

Apart from their absorption in other regions, all the compounds indicated strong specific absorption in the 9.8-10μ region, thus making infra-red absorption study in this region a highly reliable detecting tool for the existence of cyclopropane ring.

This investigation should be of considerable importance in the study of terpenic materials containing such a system. It has been applied in the case of sandalwood oil with interesting results. This should provide an infallible method for assessing the quality of sandalwood oil and for detecting any adulterant. (*Abstract*)

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